

**EXPERIMENTS CONCERNING THE MOLD MATERIALS USED IN THE
PRODUCTION OF THE COPPER INGOTS FROM THE LATE BRONZE AGE
SHIPWRECK EXCAVATED AT ULUBURUN, TURKEY**

A Thesis

by

THOMAS SCOTT LARSON

Submitted to the Office of Graduate Studies of
Texas A&M University
in partial fulfillment of the requirements for the degree of

MASTER OF ARTS

August 2009

Major Subject: Anthropology

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ABSTRACT

Experiments Concerning the Mold Materials Used in the Production
of the Copper Ingots from the Late Bronze Age Shipwreck Excavated at
Uluburun, Turkey. (August 2009)

Thomas Scott Larson, B.A., University of San Diego

Chair of Advisory Committee: Dr. Cemal Pulak

Underwater excavations of a Late Bronze Age shipwreck at Uluburun, Turkey recovered a combined 475 oxhide and plano-convex discoid copper ingots. While the hoard of ingots excavated at Uluburun brings the total number of copper ingots from the Late Bronze Age to over 1000, interestingly, only one ingot mold from that period has been identified. Scholars have speculated over the means behind the creation of these ingots for decades, but with a relative absence of archaeological molds the most promising method of reaching any conclusions as to the types of molds used in antiquity seems to be experimentation.

Experimental archaeology, has, in recent years been responsible for many breakthroughs in how the past is viewed. In the face of an overwhelming disparity of copper ingot molds from the Late Bronze Age, trials designed around testing different mold materials and casting techniques have the potential to determine, with relative certainty, how copper ingots were cast over 3000 years ago.

This thesis examines the possible materials used to create copper ingot molds through a study of their prevalence in antiquity and also details experiments in which these materials were used, in concert with different casting techniques, to create copper ingots. The results of these experiments are combined with analyses of the Uluburun ingots in an effort to bring some closure to the debate surrounding copper ingot molds in the Late Bronze Age.

DEDICATION

For my wife, the beautiful Dr. Heather Larson

ACKNOWLEDGEMENTS

Now that this project is at an end, and I have time to give pause and reflect on it, I realize that there are far too many people, other than me, responsible for its completion. Only with their contributions would these experiments have been possible. With only these scarce few lines to give thanks and acknowledge their assistance and support, I am sure I will leave someone out. So, I apologize in advance for any people I may forget in the following paragraphs.

Even though words cannot express the debt of gratitude I feel for her unyielding support and companionship since the day we met, I must thank my wife, Heather. Without her, I am quite certain these words would have never been written.

Professionally, I cannot thank Dr. Cemal Pulak enough for first taking a chance on me and allowing me to handle the Uluburun ingots for two summers in Turkey and then deeming me worthy to take on a thesis examining their production. Thank you, Cemal. I am forever in your debt. You guided me through this and this thesis is yours as much as it is mine.

Dr. Angie Hill-Price, thank you for taking on a student with a project outside the traditional realm of Manufacturing and Mechanical Engineering Technology. Without your assistance both in the analysis of my samples and their processing, this project would have most certainly stalled out.

Jim Jobling, thank you for helping me build and later store an unstable and dangerously hot-burning furnace on the grounds of the Conservation Research

Laboratory at the Riverside campus of Texas A&M. I am sorry for the mess I left behind.

To the Kennecott Utah Copper Corporation, thank you for providing me with enough impure copper to conduct these experiments.

Finally, I would like to thank the faculty, staff and students in the Anthropology Department at Texas A&M during my time in College Station. You made a kid from the west coast feel at home, deep in the heart of Texas.

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CHAPTER I

INTRODUCTION

Over the summers of 1984 through 1994 a team of archaeologists representing Texas A&M University and the Institute of Nautical Archaeology excavated a Late Bronze Age shipwreck at Uluburun, near the modern city of Kaş, Turkey. The wreck, discovered by a Turkish sponge diver, lay in approximately 50 m of water off southern Turkey (Pulak 1998: 1-2). Radiocarbon dating of items such as olive pits, leaves, brushwood, and brush dunnage, combined with dendrochronological dating of a piece of firewood among the cargo, suggests the ship sank in the last quarter of the 14th century BC (2000: 137; 1998: 214). The artifact assemblage, too, seems to support an approximate date for the sinking of this ship near the end of the 14th century BC (Bass 1986: 270). The cargo, which may have royal or elite origins, contained, among other things, Cypriot and Syrio-Palestinian pottery, glass ingots, ebony longs, hippopotamus teeth, a partial elephant tusk, ostrich eggshells, terebinth resin in Canaanite jars, one ton of tin ingots, and approximately 10 tons of copper ingots (Pulak 2000 137). Of particular interest are the 10 tons of copper ingots, as they represent the single largest group of copper ingots from the Late Bronze Age. This group comprises the bulk of ingots recovered from this period. The ingot hoard consists of 354 “oxhide” ingots, and 121 plano-convex discoid, or “bun,” ingots (Pulak 2000 137; 2001: 18).

This thesis follows the style and format of *Historical Archaeology*.

The Copper Ingots from the Uluburun Shipwreck

An extensive metallurgical study of the copper ingots from the Uluburun shipwreck was published by Hauptmann, Maddin and Prange in 2002. In addition to the observations I have made concerning the physical characteristics of the copper ingots during my two summers studying them with Michael Jones and Cemal Pulak at the Museum of Underwater Archaeology in Bodrum, Turkey, the Hauptmann et al. article will serve as the primary source pertaining to the metallurgic structure of the ingots, and any conclusions about their origins based on scientific analyses. I will attempt to summarize the main aspects of their article, as they relate to this paper.

The most pertinent aspect of Hauptmann et al.'s study of the Uluburun copper ingots to this paper is the attempt to extrapolate information regarding the mold materials used to cast them based on a careful study of their grain structure (Hauptmann et al. 2002: 3). In order to examine the grain structure of the Uluburun ingots, the authors took core samples from the centers and, in one instance, from the corners of select copper ingots using a tubular drill with a 6 mm inner-diameter. Preparation of the specimens for microscopic examination consisted of sectioning, polishing, and etching them with a 1:1 mixture of NH_4OH and H_2O_2 (Hauptmann et al. 2002: 3). The study revealed two metallographic characteristics common to almost every sample examined: constant porosity throughout each ingot and the uniform presence of different phases and compounds in the matrix (Hauptmann et al. 2002: 4). On average, the ingots contained as much as 20% porosity by volume. The cause of this feature was the effervescence of

gases such as oxygen, carbon monoxide, carbon dioxide, water vapor, and sulfur dioxide in the liquid copper as it cooled after casting (Hauptmann et al. 2002: 4). According to the authors, high porosity such as this is common with blister ingots originating from Cyprus. High porosity in these types of ingots is due to the oxidic nature of the ores used, which inhibit the degassing of sulfide inclusions in ores such as chalcopyrite (Hauptmann et al. 2002: 5).

The authors stated that the grains of the Uluburun ingots were equiaxed to a size of 3 on the ASTM (American Society for Testing Materials), which means that they were between 50-120 μm (Hauptmann et al. 2002: 6). The equiaxed character of the copper grain structure alludes to the slow cooling of the ingots after casting. Hauptmann et al. noted that there was “no columnar characteristics, as would be expected if the cooling had occurred quickly in a mold material with a thermal conductivity greater than that of stone or earth” (2002: 6). Although that conclusion could have been assumed, given the lack of technology in the Late Bronze Age, the observation does provide the conclusive scientific data to back the supposition that Late Bronze Age copper ingots were not cast in metal molds. Additionally, Hauptmann et al.’s article admittedly cannot argue against the use of sand or clay molds in producing the Uluburun copper ingots, although no traces of quartz, clay, or grains of other materials were observed adhering to the surface of the ingots (2002: 18). Given this information, it was decided that three materials readily available to Late Bronze Age smelters would be investigated in this study: clay (earth), stone, and sand. The experiments detailed in this paper, using the above mentioned materials, aim to gain insight into the details of casting copper ingots

in the Late Bronze Age and determine the type of molds in which they were cast.

Hauptmann et al. (2002: 17) confess that the technology of ingot production has hardly been investigated and is consequently an area of inquisition that needs the attention of scholars.

The authors also made interesting observations that should put to rest some of the debate concerning the production of copper ingots in the Late Bronze Age. They determined, contrary to some theories, that copper ingots were not created in a single-step process by smelting copper ore in tap furnaces, but rather involved intermediate refinement, remelting and then were poured from a crucible (Hauptmann et al. 2002: 12-17). The relatively high purity of the copper used to create the Uluburun ingots demonstrates this supposition, as impurity in the ingots is less than 1%. This number is slightly misleading, however, as the composition of each ingot is not necessarily 99-100% copper, due to non-homogeneous inclusions of slag and gas pores (Hauptmann et al. 2002: 13).

Prior to this study, some researchers had posited that plano-convex discoid ingots were the result of copper left in the bottom of a furnace after the smelting of copper ore (Tylecote 1976: 159, 170; Wheeler et al. 1975: 32). However, Hauptmann et al. have demonstrated that the formation of bun ingots is contrary to this manner, that they are the result of many refinements and poured from a crucible in the same manner as oxhide ingots. The reasoning behind this claim lies in the analysis of slag inclusions found in the Uluburun ingots. Hauptmann et al. have concluded through their study that the small bits of slag inclusions, given their angular nature, were in a solid state at the time of

casting, since globular slag inclusions are more indicative of liquid slag at the time of casting. Were ingots created in the smelting furnace (as some have postulated for the bun ingots), or poured directly from a tap smelting furnace, the slag inclusions in the molten copper would have been liquid at the time of casting. Since the slag inclusions were solid, Hauptmann et al. (2002: 17) construe this to be evidence that the copper was melted in and poured from a crucible, where furnace temperatures did not reach high enough temperatures to turn slag inclusions into a liquid.

Physical Appearance of the Uluburun Ingots

Since Hauptmann et al. (2002: 18) and Muhly et al. (1977: 357) agree that size is the only major difference between copper bun and oxhide ingots, it is important to look at the physical characteristics that are manifest in these ingots because of their composition and the molds that were used in casting them. During the summers of 2003 and 2004, Michael Jones and I, under the supervision of Cemal Pulak, elaborated on the existing catalogue for the copper oxhide ingots recovered from the Uluburun shipwreck. This task involved not only checking the if artifact numbers matched the actual artifacts, but also that their weight, dimensions, and verbal descriptions were correct. Additionally, we studied and incorporated any ingots not previously included in the catalogue of the Uluburun ingots. Since a comprehensive terminology regarding the casting features of raw materials such as ingots does not exist, we adopted certain terms

from finished cast products to describe the features of the Uluburun ingots. These terms are listed and defined in the glossary contained in Appendix B.

Given that this study attempted to replicate copper bun ingots from the Uluburun shipwreck as closely as possible, and in doing so determine the mold materials used in their production, the physical descriptions of Uluburun ingots will be limited to those features present on all ingots. Perhaps the most obvious feature of these ingots is that they possess two distinctly different surfaces. The observation that copper ingots have two such conspicuous surfaces indicates that only one surface of the ingot was in contact with a mold, not both. Furthermore, that one of the surfaces is smooth and retains the shape of the mold in which it was cast, while the other is irregular and bubbly, indicates that all of the copper ingots, bun and oxhide alike, were cast in open molds. While this is a seemingly obvious statement to make given the differences in the two sides of copper ingots, the idea that Late Bronze Age copper ingots were cast in open molds was relatively new when it was echoed by Bass (Bass 1961: 272) in his examination of the copper oxhide ingots recovered from the Cape Gelidonya shipwreck. This simple observation is crucial, because this basic fact determines the form and material of the molds used to cast ingots in the experiments of this thesis.

With respect to copper ingots from the Late Bronze Age, molten metal poured and allowed to cool in an open mold creates smoother, yet porous sides, and a bottom surface. The bottom surface is defined as the mold surface, since it is permanently in contact with the mold during casting. Identification of the mold surface of an ingot is contingent on two key features: its relative smoothness, when compared to the rough

upper surface of the ingot, and numerous gas pores, which are defined as holes in the surface of an ingot caused by gasses within the ore released during the casting process.

No ingot possesses a completely smooth mold surface; all are marked by swells of varying sizes. Swells are deformations of an ingot's mold surface due to the pressure of molten metal moving or displacing the sand of a mold, or causing spalling, and/or irregularities in a stone or clay mold. Before experimentation, I correctly anticipated that casting in sand molds and repeated castings in reusable molds of stone and clay would result in determining the mechanism behind the creation of swells. Successive castings seem to have revealed that swell prominence decreases with each successive casting, which is contrary to previously conceived notions regarding swell progression on mold siblings and will be discussed in greater depth later in this paper. Pulak asserts the presence of mold siblings among the bun and pillow-shaped small oxide ingots of the Uluburun wreck through a comparison of ingot sizes, shapes and irregularities as well as matching the swells on their respective mold surfaces (All of the Uluburun bun ingots were examined by Cemal Pulak and Samuel Lin 2001 and 2002 in order to determine the presence of mold siblings within the assemblage. The results of this study are not yet published.).

In order for swells to be replicated in an ingot cast in a sand mold, a template of some sort would have to have been used in preparing a new sand mold for each ingots, since the sand mold would have been destroyed with each casting. Conversely, for matching swells to occur on ingots cast in reusable molds, there would have to be some progressive spalling of stone or clay when successively casting ingots in the same mold.

This progression of spalling would manifest itself in gradual changes to the swells on the mold surfaces of mold siblings.

Extensive, coarse blistering is the principal diagnostic feature of the rough upper surface of a copper ingot. Blisters are here defined as shallow blows (round or elongated holes generated by trapped gases) covered over with a thin film of metal. As expected, blistering makes this surface irregular and rough to the touch, giving it an almost gravelly appearance, hence the term, “blister ingot”, by which these ingot types are known in metal smelting.

The edges of the rough surface of a copper ingot also bear some unique diagnostic features. The outermost edges of the rough surface is marked by a cooling ridge, or a low, narrow elevation where molten metal of an ingot first solidifies, thereby creating a barrier for molten metal to pile up against. Cooling ridges seem to vary in size and prominence, but they are still a feature common to all of the copper ingots from the Uluburun shipwreck. A cooling ridge is always accompanied by a shrink depression, which is a long, continuous, narrow, concave surface near the outer edges of the rough surface of an ingot. This combination of features are formed due to the narrow outer edges of an ingot cooling faster than the large inner areas. This results in the formation of large copper crystals. Contrarily, the slower cooling metal near the center of the ingot forms smaller, more compact crystals. The discrepancy between the sizes of crystals results in a concave depression near the edge of an ingot. Occasionally, a shrink depression may be so slight that it is only noticeable through a decrease in the severity of

blistering near the edges of the rough surface, rendering a comparatively smoother border to the rough surface of an ingot.

Experimental Archaeology in Casting Copper Ingots

Despite the relative abundance of copper ingots from the Late Bronze Age, little definitive knowledge exists about their production. After surveying much of the literature pertaining to the copper ingots from the Late Bronze Age, as well as investing four months over two summers cataloging and studying the Uluburun ingot hoard, it became apparent that the only real means of getting at the root of the questions pertaining to these artifacts, and hopefully reaching meaningful conclusions, was through experimental archaeology. Indeed, experiments designed to test the various mold materials that may have been used to cast copper ingots in the Late Bronze Age is essential to determining how those ingots were produced. Experimental archaeology, in the case of this study, will hopefully remove some of the guesswork involving the casting process of these ingots and replace it with experimentally reproduced material evidence.

After becoming intimately acquainted with the Uluburun copper ingot collection through working with them for two summers in the Museum of Underwater Archaeology in Bodrum, Turkey, it became startlingly apparent that no matter how many times one runs hands over an ingot, how many times one pours over the smallest of surface details, authoritative information pertaining to how they were created could

only be ascertained by creating the ingots oneself. Experiments designed to melt copper and cast it into ingot form have the potential to shed light on several aspects of Late Bronze Age ingot production. Specifically, the experiments proposed and detailed in this thesis should offer insights as to the mold materials used to create the ingots, the number of pours of molten copper used to create ingots, the production of mold siblings, and the processes behind the creation of features, such as swells and blisters, common to all copper ingots from this period. Ultimately, the products of the casting experiments detailed in the following paper will be compared to archaeological samples from the Uluburun collection of ingots with the goal of determining definitively the mold material used to cast the Uluburun ingots. Additionally, the information gained from these experiments will provide a strong foundation for future comparisons with other copper ingots from the Late Bronze Age and hopefully will allow researchers to determine the mold materials used to cast all ingots analyzed from the Late Bronze Age, not just those recovered from the ship wrecked at Uluburun.

CHAPTER II

LITERATURE REVIEW

Copper Ingots from the Late Bronze Age

Evidence found in paintings on the walls of the tomb of the Late Bronze Age Egyptian official, Rekhmire, led scholars to associate copper oxhide ingots to sea-born trade involving mostly ancient Syrians (Bass 1967: 7; Pulak 2000 138; Wheeler et al. 1975: 32-33). Research dedicated to the study of copper oxhide ingots comprises the majority of publications concerning ingots from the Late Bronze Age, while less is known about plano-convex discoid ingots. Both ingot types were transported on the shipwrecks excavated at Uluburun and Cape Gelindonya, though oxhide ingots outnumbered the plano-convex discoid type nearly three to one. Despite the differences in shape between the ingot types found on the Uluburun wreck, there does not appear to be any disparity among the ingots with respect to casting techniques or purity of copper (Pulak 2000 140, Hauptmann et al. 2002: 17), indicating that all were produced using similar, if not identical techniques.

Early research into the production of copper ingots suggests production by smelting of raw copper ore (Tylecote 1981: 94) in blast furnaces fueled with charcoal (Forbes 1964: 18). It was also commonly held that a single ingot was the product of the ore from a single mine (Wheeler et al. 1975: 32). Tylecote (1981: 89), when referring to a furnace proposed to be responsible for the production of oxhide ingots further

suggests, “Ore is directly reduced in one stage yielding copper metal”. This belief, however, proved to be somewhat problematic, as archaeologists have yet to find furnace remains at ancient copper mines. A classic example of this paradox can be seen in the fact that there have been no ingot molds recovered from the copper mines at Timna, Israel, and no furnaces at Ras Ibn Hani, Syria at the site of the only known oxhide ingot mold (Maddin 1988: 179; Lagarce et al. 1983: 249). Given the lack of furnaces at mining sites, mines and smelting centers, it is conceivable that copper production was a multi-stage endeavor, where ancients mined raw copper at one location, then shipped it to and refined it at a secondary location.

Indeed, recent research has revised many of the initial theories related to the smelting of copper for the production of ingots. Tylecote (1992: 35, 37) has changed his position to support the remelting of smelted copper, minus slag, in order to cast copper oxhide ingots. Hauptmann et al.’s (2002: 12-17) recent examination of the Uluburun ingots supports these revised theories, as their study revealed that the copper used to make the Uluburun ingots was, in fact, far more pure than any ore source and, therefore, had to have been the result of a multi-step refining process.

Nevertheless, despite extensive studies delving into the production of copper ingots, there are no definitive conclusions relating to the casting materials for copper ingots in the Late Bronze Age. Based on visual inspection of the Uluburun ingots, Pulak (2000: 141) suggests that at least some of the “two-handled” oxhide ingots were cast in sand molds due to the presence of fins on the sides of the ingots, while the plano-convex discoid and “pillow” ingots were cast in clay or stone molds. The presence of fins on the

side of an ingot most likely results from molten copper seeping into the sides of a mold and then solidifying, creating a fin-like protrusion. It would be difficult for such a feature to be formed on ingots cast in molds of stone or clay, because the hardness of those two materials would make seepage unlikely. Sand molds, however, could easily allow for the formation of fins if the sides of the mold were to crumble during casting. Yet, the metallurgic study conducted by Hauptmann et al.'s study suggests, based on microscopic examination of the ingots' crystalline structure, there is no difference in casting techniques among the Uluburun ingots (2002: 18). Some scholars have suggested that bun ingots were created using the cooled and hardened, left over, material in the bottom of furnace from the smelting of copper for larger ingots (Tylecote 1975: 159, 170; Wheeler et al. 1975: 32). Others argue for casting bun ingots in molds independent of a furnace (Merkel 1986: 256; Rothenberg 1990: 54; Tylecote 1992: 37). While most scholars agree ancients cast oxhide ingots outside of a furnace (Tylecote 1976: 170; 1981: 90; Wheeler 1975: 32) the types of molds in which they were cast is still the subject of much discussion.

With so many differing theories surrounding the production of copper ingots, it is clear further research is needed in order to bring some uniformity to the discussion. Given the technology available to copper smelters in the Late Bronze Age, there are only three realistically viable mold materials in which ingots could have been cast: clay, sand, and stone. Some researching this subject argue for the use of sand molds (Bass 1967: 70; Merkel 1986: 259; Tylecote 1981: 94; Wheeler 1975: 32), others for the use of clay molds (Tylecote 1981: 94; 1992: 37; Rothenberg 1990: 54), still others support stone

mold use (Craddock 1997: 5; Lagarce 1983: 277-79; Tylecote 1992: 38-40). With so many differing, yet informed, opinions on the matter, something more than research needs take place to settle the debate surrounding the molds used in the production of Late Bronze Age copper ingots.

A review of evidence exploring the different possible materials used to cast ingots in the Late Bronze Age should be undertaken before any experimentation can take place. Archaeological evidence was considered from the Early and Middle Bronze Ages, in addition to what is known from the Late Bronze Age. The reasoning for this is simple: casting technology known to copper smelters in the Early and Middle Bronze Ages was likely handed down through generations of metal smelters and became well established in the Late Bronze Age. Since the only materials from these epochs capable of withstanding the temperatures of molten copper were clay, earth (sand), and stone, an extensive search for all examples of such materials was carried out. Additionally, since the technology level of the copper producing tribes of central Africa was probably not more significantly advanced than that of cultures of the Bronze Ages, an examination of the ethnographic and archaeological evidence of molds used by those tribes was also undertaken in order to assist these experiments.

Previous Experimentation

Experimental archaeology has been in existence in some form for over a hundred years. In that span of time, many have conducted experiments into the mechanisms

behind copper production, not only in the Late Bronze Age Mediterranean, but in the New World as well. It is important to understand what work has been done previously and how that research can help direct these experiments into the various potential mold materials used to cast copper ingots in the eastern Mediterranean some 3300 years ago.

Despite much interest in copper ingots, Craddock et al. (1997) are the only researchers to have conducted experiments focused directly on determining the legitimacy of stone as a material used for the use of casting copper ingots. With the discovery of the limestone mold at Ras Ibn Hani, which happens to be the sole representation of a stone mold for the casting of copper oxhide ingots, Craddock, Freestone, and Dawe performed trials designed to test whether or not limestone was a suitable material for the casting of copper ingots. Their experiments were an attempt to either prove or disprove the use of the Ras Ibn Hani artifact as an actual mold used in the production of copper oxhide ingots in the Late Bronze Age.

Craddock et al.'s experiments are especially important, because prior to the discovery of the Ras Ibn Hani mold, it was commonly believed that limestone, while abundant in the region, was not a suitable mold material due to its thermally unstable nature when heated to temperatures above 900°C (Craddock et al. 1997: 1). For their casting experiment, Craddock et al. used limestone obtained from Gozo, a small island off the coast of Malta. This specific limestone was chosen because of its heat resistant nature. In fact, Dawe, while on Gozo, discovered that this particular type of limestone (apparently only available from one mason who obtains it from one quarry) is commonly used in the construction bakers' ovens on Gozo (Craddock et al. 1997: 4). What

seemingly made this type of limestone suitable for exposure to high temperatures was its tighter grain structure and fewer fossil inclusions than seen in more commonly found types of limestone. In fact, the Gozo stone quite closely resembles the stone used to carve the Ras Ibn Hani mold (Craddock et al. 1997: 4).

Craddock et al. carved into a 360 mm by 275 mm by 95 mm block of the Gozo stone, a half-scale negative of an oxhide ingot at a depth of 15 mm. No pretreatment of the mold occurred during casting, with the exception of heating with a blow torch to dry the contact surface of the mold. Molten copper was poured into the mold directly from a crucible, and an ingot weighting 2.2 kg, with a thickness between 12 and 15 mm was cast (Craddock et al. 1997: 4). The mold survived the casting intact, but some of the limestone on the contact surface spalled and adhered to the ingot (Craddock et al. 1997: 5).

John Frederick Merkel, in his work concerning copper smelting in the early 1980s produced several copper ingots in molds of both sand and clay (Merkel 1982: 335) as well in the bottom of a shaft furnace (Merkel 1982: 277).

Merkel used a fired clay mold made from mixture of kaolinite and sand to cast two copper bar ingots. The clay mold broke on its third use, not from casting, but rather mishandling (Merkel 1982: 278). Microscopic examination of cut sections of the ingots revealed the top surface contained a proportion of columnar grains and the middle grains were more equiaxed; cuprous oxide was evident at grain boundaries” (Merkel 1982: 278-279). The similarities between Merkel’s experimental copper bar ingots and those

from antiquity is important and useful, as it lends credence to the idea that clay is a viable mold material for casting larger plano-convex discoid and oxhide ingots.

Merkel also cast a full-sized oxhide ingot in a sand mold during his experiments. The ingot weighed 25.1 kg and was 50 cm by 23 cm and 4 cm thick. The only observations offered by Merkel concerning the oxhide ingot was that its “top surface had a rough appearance and exhibited a copper oxide crust” (Merkel 1982: 270). The oxhide ingot was sectioned for examination, but its grain structure is not discussed. While Merkel did create a number of ingots during his experiments, there is little discussion of their makeup, as he was primarily concerned with the processes behind rendering copper from its ore, as well as furnace construction and operation used in that process.

Sven Van Lokeren experimented with sand as a viable mold material for the casting of copper oxhide ingots. In the experiment, one copper oxhide ingot was cast by melting 30 kg of copper in a gas furnace and then pouring it from a crucible into a mold created by impressing a wooden pattern into sand (Van Lokeren 2000: 275). Van Lokeren does not discuss the type of sand used in the mold, nor if it was bonded with any other material or contained any moisture to help it hold its shape. Furthermore, the resulting ingot is not discussed in any detail, other than it was reheated and smashed with a sledge hammer. Van Lokeren does, however, attribute the ease in which the ingot breaks to its makeup of 20-30% gaseous inclusions (Van Lokeren 2000: 275), but does not attempt to explain the origins of the porosity as being from the gases effervescing from the cooling metal or evolving from the sand mold.

Although not concerning copper ingots, G. Clement Whittick did conduct informative research concerning the amount of pours used to create Roman lead ingots. Whittick's research in this area is noteworthy here, because little to no information has been published about the possible separation lines found on their edges. Whittick's experiments demonstrated that although Roman lead ingots appeared to have been cast in multiple pours, the striations on their sides were instead the result of lead cooling and hardening as it comes in contact with the mold surface (Whittick 1961: 108). The freshly poured lead, as it comes in contact with the mold, cools inward and upward. Thus as more lead is poured into the mold, it will overlap and cover the existing cooled lead, cool and solidify itself, and create a second pile of lead atop the first. This process repeats itself as long as lead is being poured into the mold. The end result is the multiple striations seen on the sides of Roman lead ingots (Whittick 1961: 108).

Others have also conducted experiments on producing copper objects, but their work was more focused on how ancients extracted copper from its ores. Cushing published work in 1894 detailing his efforts to render metallic copper from ore and then anneal it much like the Hopewell and Zuni cultures from North America (Cushing 1894). Coghlan, too, conducted experiments in the late 1930s in an attempt to ascertain how ancients may have discovered the possibility of obtaining metallic copper from ore. His efforts examined the potential of the "hole in the ground" furnace as the earliest means of extracting metallic copper, as well as the pottery kiln. Coghlan was unsuccessful in his attempts to reduce pieces of malachite using the "hole in the ground,"

technique and thus concluded that the discovery of metallic copper must have come from the use of pottery kilns (Coghlan 1938: 106-108).

While much of the previous research into copper ingot production seems focused on gaining insight into furnace construction and operation, or the most basic means of extracting metallic copper from ore, very little is dedicated to the study of the mold materials in which copper ingots were cast. In fact, only Craddock et al. (1997), Von Lokeren (2000), and to a very small extent, Merkel (1982) dedicated any time to the study of ingot molds. Collectively, all of these efforts provide a framework for further research by highlighting the various potential mold materials in which ingots could have been cast. However, aside from cursory investigations of limestone (Craddock et al. 1997), sand (Van Lokeren 2000), and clay (Merkel 1982) as potential materials for ingot molds, no effort has been made to compare the products of these mold types with each other or against an archaeological collection. In fact, where the above mentioned research falls short is in this realm; the research confirms that these materials can be used to cast copper objects, but makes no attempt to validate their use in the Late Bronze Age with respect to the casting of copper ingots. If compelling arguments are to be made as to what materials were used to cast copper ingots in the Late Bronze Age, ingots have to be cast in molds of all three materials and the products compared to each other, as well to an existing collection of archaeological ingots. These comparisons should be done not only on the macroscopic level, but on the microscopic level as well.

Clay Molds

Pulak (Pulak 2000: 141), citing the presence of mold siblings among the “pillow” ingots from the Uluburun collection, suggests the use of reusable clay or stone molds for ingot casting. Although clay ingot molds from the Late Bronze Age are nearly non-existent, that are found in Early and Middle Bronze Age contexts (Dever and Tadmor 1976; Rothenberg 1990; Levy et al. 2002; Weisgerber 2003). Moreover, the evidence for the use of clay molds in casting copper ingots by the tribes of Africa’s Copper Belt indicate that an investigation of clay as a possible mold material for the casting of ingots in the Late Bronze Age is prudent and warranted.

Excavations at site 30 of the 10th-century BC settlement at Timna, Israel have revealed 21 clay ingot mold fragments (Rothenberg 1990: 54). The fragments were heavily sintered and appear to have been made with slag-tempered clay, but exhibited no evidence of slagging (Rothenberg 1990: 54). The absence of evidence for slagging indicates that these molds were used for the casting of relatively pure copper, which is indicative of the quality of copper ingots from the Uluburun shipwreck. Based on their shapes, Rothenberg (1990: 54) purports the fragments were used to cast plano-convex discoid ingots of oval or rectangular shape. Rothenberg suggests that other mold fragments have been found but were dismissed as furnace fragments due to the similarities between the two refractory materials (Rothenberg 1990: 3).

Based on this reasoning, there is also a good chance that other clay ingot molds have not been reported because it is likely that archaeologists were not looking for them

or were unable to identify them. Some researchers initially hypothesized that circular plano-convex discoid ingots were obtained from left over copper that had pooled in the bottom of smelting furnaces (Tylecote 1976: 159, 170; Wheeler et al. 1975: 32), and were not cast in molds external to the furnace. Since such a reasoning would not require the use of circular plano-convex discoid ingot molds for the production of bun ingots, it is conceivable that some refractory materials found at Late Bronze Age smelting sites were thought to be furnace remnants and not examined to evaluate their potential for use as clay ingot molds.

Both the Early Bronze Age sites of Hamr Ifdan and Barqa al Hetiye have yielded a significant amount of mold fragments for casting bar-ingots. The molds are made of clay, contain a groove with a rounded profile in their upper surfaces into which copper was poured, and are brick-shaped (Weisgerber 2003: 83). Unfortunately, a detailed study of the composition of these molds has yet to be published, which means nothing more of their material makeup can be ascertained. The discoveries of these molds are significant despite the lack of published extensive examination and research, as they seem to establish the use of clay as a mold material for casting copper ingots in the Early Bronze Age, well before the creation of the ingots recovered from the shipwreck at Uluburun.

Sealed by an earthquake some time between 2700 and 2200 BC, Khirbet Hamra Ifdan is the largest metalworking center of the Early Bronze Age and offers superb preservation of finds (Levy et al. 2002: 425). Finds from the site include crucible fragments, ceramic (clay) molds for both ingots and tools, and copper ingots. In total, 60 copper ingots and 361 ingot mold fragments have been recovered from this site (Levy et

al. 2002: 425-429). Interestingly, the ingot mold fragments were found in a refuse pile and all appear to have been broken to retrieve the metal objects cast therein (Levy et al. 2002: 433). Since the molds appear to have been broken and discarded, it is unlikely they would have produced any mold siblings from this site. This is contrary to findings from plano-convex discoid ingot production in the Late Bronze Age, as evidenced by the presence of many mold siblings among the findings from the Uluburun shipwreck. The fact that the molds from Khirbet Hamra Ifdan had to be broken to retrieve the metal, thus rendering them a one-use item, can be construed as evidence for the use of a non-stick agent such as ash, or more durable molds altogether, in the Late Bronze Age. The practice of lining molds with ash was common to the Yeke and Sanga tribes of central Africa (Bisson 2000: 96, 99). Although, it is conceivable that these molds remained in use for the casting of successive ingots and were not broken until an ingot could not be retrieved using conventional methods.

In addition to the discarded clay ingot molds at Khirbat Hamra Ifdan, archaeologists recovered numerous clay molds used to cast other objects, such as pins and axes. These molds appear to be made of fired clay and, in the case of the pin molds, were used to produce multiple items at one time. Given the similarity in size and shape to each cavity used for casting within an individual mold, it appears each of the pin mold impressions were made from a template of some sort being imprinted into wet clay. The molds exhibit significant charring and are unbroken, indicating they were intended and multiple use items (Levy 2007: 86).

In 1974, a hoard of 21 copper ingots was found near El-Hadab, Israel. Although the hoard was discovered by archaeologists only after it was removed from its context, the ingots appear to date to the Middle Bronze Age (Dever and Tadmor 1976: 164). Investigations of the area purported to have yielded the hoard of ingots include a fragment of a small baked clay mold, which Dever and Tadmor (1976: 163) believe could have been used to cast ingots like those of the hoard.

Although little research has been done on the clay ingot molds recovered from the Early and Middle Bronze Ages, their existences supports the idea that ancients did, in fact, use and know how to produce clay molds for the casting of copper ingots. Furthermore, the continued use of clay molds from the Early Bronze Age and into the Middle Bronze Age allows for the speculation that this was indeed a technology that was used and passed down from one generation to the next, which makes it entirely possible that casting copper ingots in clay molds was a solidly established technology, well known to copper smiths in the Late Bronze Age.

Yet, despite finds from the Early and Middle Bronze Ages, only one site, Timna, yielded evidence for the use of clay ingot molds from the Late Bronze Age. Tylecote attributes this lack of evidence to the idea that clay molds are too fragile, and therefore have not survived the 3300 years to modern times (Tylecote 1992: 40). However, this argument can be easily refuted by the presence of clay crucibles and tuyeres (Knapp et al. 2001: 206-208) dating to the Late Bronze Age, not to mention the finds discussed above from the Early and Middle Bronze Ages. Yet, it is highly dubious that molds from

the Early and Middle Bronze Ages would survive until modern times, while Late Bronze Age clay ingot molds happen to disappear altogether.

Adding to the mystery of why very few clay ingot molds have been recovered from the Late Bronze Age is the presence of many clay molds recovered from copper smelting sites in Central Africa (de Maret 1995; Bisson 2000: 103-20), some dating to as early as the ninth century AD (Bisson 1976: 223). Although the molds recovered from the Congo region cannot be placed on a timeline of similar magnitude as those of the Late Bronze Age, the reality is that clay materials do survive from antiquity. If clay ingot molds were used to cast ingots in the Late Bronze Age, evidence of their existence should have survived in one form or another. Although, it is conceivable the inevitable destruction of such molds after repeated usages has prevented them from being easily identified. A destroyed bun or oxhide ingot mold would not bear the same diagnostic features as a production mold used to create, for example, a small figurine, making the former much more difficult to identify. Additionally, clay molds are made from similar refractory materials to those used in furnace linings and tuyeres. As such, they could be mistakenly identified as the latter, more common items, since it is not likely that clay refractory remnants were being examined by researchers believing plano-convex discoid ingots were formed from molds external of the smelting furnace. Nonetheless, it should be remembered that the absence of evidence does not necessarily equate to evidence of absence, especially in the face of heavy usage of clay ingot molds during the Early and Late Bronze Ages, and in medieval Africa.

The Savannah kingdoms of Central Africa were once known for their copper smelting (De Maret 1995), despite a lack of technology by modern standards.

Investigations into the seemingly antiquated ways in which they smelted and cast copper has the potential to yield information regarding eastern Mediterranean ingot casting in antiquity. Finds from the site of Naviundu, near the Zambian-Congo border can be dated to the mid-fourth century AD and are commonly associated with the inception of copper metallurgy in the region (Bisson 2000: 115). Clay ingot mold fragments from the site of Kipushi have been dated as early as the ninth century and are the earliest known in the region (Bisson 1976: 223). This information, coupled with ethnographic evidence from the late 19th century (Arnot 1889: 238), indicates that copper smelting in the region has been in continuous practice for more than 1500 years, clay as a mold material for ingots has been used for just over a thousand years, and both have remained relatively unchanged over that great span of time. Given this knowledge, it is possible that an analysis of the molds used by the tribes of Central Africa during that time to cast copper ingots will yield important analogues to the copper casting techniques and mold materials used in the Mediterranean during the Late Bronze Age.

As noted above, archaeologists have solid evidence that metallurgy in Central Africa dating to the mid-fourth century AD (Bisson 2000: 115). Despite the early date for copper metallurgy, clay mold usage cannot be securely dated until around AD 860 at the site of the Kipushi mine in Zaire, near the Zambian border (Bisson 1976: 223). The Kipushi mine was one of the largest in the eastern Zairian Copper Belt (Bisson 1976: 210). Molds from this site were used to produce un-flanged, H-shaped copper ingots

(Bisson 1976: 427), which do not appear later than the 12th century AD (Bisson 2000: 120). Many clay molds have been recovered there, throughout the habitation sites as well as near the metal production centers (Bisson 1976: 218). In total, 42 clay ingot mold fragments were recovered from Bisson's excavations at Kipushi in the 1970s (Bisson 1976: 427). The largest of these fragments would have produced an ingot roughly 17 cm long and 1 cm thick (Bisson 1976: 427).

In addition to the mold finds at Kipushi, miniature copper ingots, generally referred to as HH-type croisettes, common to the Upemba basin and Kamoia in the 16th and 17th centuries, were cast in rectangular terracotta blocks. Each of these blocks was designed to cast four to six ingots simultaneously (de Maret 1995). Finds such as this speak to the widespread usage of clay-type materials serving as a means to cast copper ingots in Africa. The popularity of such a material among copper producing peoples cannot be discounted when it comes to experiments into the production of copper ingots from the Late Bronze Age.

Refractory Materials

With the relative absence of clay ingot molds from the Late Bronze Age, one must look to clay molds used for items other than ingots as well and clay items relating to metallurgy, such as furnace linings, tuyeres, and crucibles, when deciding how to best replicate a clay ingot mold for use in casting experiments. By analyzing the refractory materials used in the Late Bronze Age, as well as earlier and later periods, one can get a

sense of how the ancients would have likely constructed a mold that was to be repeatedly subjected to, and withstand, the high heat of molten copper. After examining the means by which many types of refractory items were manufactured, it can be reasonably assumed that materials common to furnace linings, tuyeres, crucibles, and non-ingot clay molds from the Late Bronze Age and other periods may well have been incorporated into the manufacture of clay molds for casting copper ingots.

One seemingly common theme with respect to the clay used to produce ancient refractory items is that it is similar to modern potter's clay (Freestone 1989: 156). However, clay alone would not be sufficient for producing an ingot mold, and some sort of temper would be necessary to provide it with refractory qualities. Typically, furnace linings and tuyeres from the Late Bronze Age were constructed of clay tempered with chopped grass, ground tap slag, and ash added to increase resistance to the high temperatures encountered when smelting copper (Freestone 1989: 156).

Perhaps more closely related to clay ingot molds are crucibles and clay investment molds from the Late Bronze Age, as they too had to withstand the temperatures to melt copper and the thermal stresses associated with being in contact with molten metal. Crucibles of the period were constructed of unfired clay containing large quantities of crushed rock and other mineral fragments (Freestone 1989: 157-58). In her study of crucible fragments from ancient Britain, Hilary Howard that noted her specimens contained 60-80% quartz particles (Howard 1983). This indicates clay is not the primary component of these artifacts, but was rather the material used to bind more thermally stable inclusions. The high filler content noted in the crucibles helps to

minimize shrinkage and cracking of the crucible when it is exposed to the intense heat of molten copper (Freestone 1989: 157). In addition to inclusions high in silica and other non-plastics, chopped grass, straw, and other vegetal matter have a long history of use in Middle-Eastern refractory materials (Freestone 1989: 159).

In general, therefore, clay molds from antiquity bear similarities to crucibles in that they are primarily made of non-plastic inclusions with clay serving as a bonding material. Usually, inclusions found in clay molds are silica, charcoal, hair, chaff, and grain (Freestone 1989: 160). The inclusions are an important component to the molds not only for their ability to prevent shrinkage and cracking, but for also making the molds porous enough to allow the gas evolved from molten copper a means of escaping without becoming too volatile (Freestone 1989: 160). In addition to allowing the discharge of effervescing gases, inclusions in clay molds increase the material's resistance to thermal shock and make the molds more dimensionally stable (Freestone 1989: 160).

Upon reviewing these four refractory items, it is apparent that the addition of some non-plastic, silica inclusion is necessary in materials intended to be exposed to the high temperatures required for casting copper. Perhaps the most common refractory inclusion mixed with clay is simply common sand, as demonstrated by the seeming ubiquitous presence of quartz/silica in furnace linings, tuyeres, crucibles, and clay molds from the Late Bronze Age. Utilizing the information provided in Freestone's analyses of refractory materials referred to above, the clay mold to be used in casting experiments for this study will be constructed from 10 pounds common potter's clay mixed with

large amounts of sand and organic materials such as grass and charcoal (roughly 40% sand and 20% organics by volume).

Stone Molds

The only mold for casting oxhide-shaped ingot mold dated to the Late Bronze Age was discovered in the north palace at the site of Ras Ibn Hani, near Ugarit in Syria. The mold dates to the 15th century BC (Lagarce et al. 1983: 279), more than a hundred years prior to the sinking of the shipwreck excavated at Uluburun. The mold is cut from a slab of limestone measuring 155 cm long, 80 cm wide, and 18 cm high. The center of the slab holds an 8 cm-deep carved cavity in the shape of an oxhide ingot. The cavity is irregularly worked (likely from tool marks used to create it), yet smooth to the touch. Lagarce et al. (1983: 277) claim the craftsmanship of the mold is fine and expertly done, similar to the cut-stone walls of the palace at Ras Ibn Hani. At one corner of the slab is a channel that leads from the edge of the slab to one of the corner projections of the carved ingot mold (Lagarce et al. 1983: 277). It is likely this channel was used either as a means of directing molten copper into the mold, or for providing a low leverage point to lever out solidified ingots. Finds near the mold pertaining to metallurgy include crucible fragments, tuyeres, a portion of an earthen oven, fragments of bone, and small droplets and pieces of copper (Lagarce et al. 1983: 277).

Curiously though, there are no copper smelting furnaces at Ras Ibn Hani, just as there are no ingot molds from one of the largest copper production sites on the Late

Bronze Age at Timna, Israel (Maddin 1988: 179). Nevertheless, the shape of an oxhide ingot carved into the surface of a slab of limestone is significant evidence and implicates limestone as a viable material for the casting of copper ingots in the Late Bronze Age. In fact, Craddock et al. (1997: 4) state unequivocally that the Ras Ibn Hani mold was used for the casting of oxhide ingots based on the presence of tiny droplets of copper surrounding the mold. They also point to the fact that the mold appears to be burned and damaged as a result of exposure to extreme heat.

Despite what appears to be unequivocal evidence for the use of limestone as a mold material for copper ingots in the Late Bronze Age, the thermal properties of limestone, namely that it becomes thermally unstable at 900°C, could lead to potentially dangerous situations, as the melting point of copper lies well above that threshold at 1083°C. When limestone is heated beyond 900°C, it breaks down into calcium oxide and emanates carbon dioxide (Craddock et al. 1997: 1). The volatility of the reaction is directly proportional to the temperatures to which the limestone has been exposed. Therefore, exposure to molten copper could result in the rapid evolution of gases that may cause dangerous spewing of liquid copper. Early casting experiments by Pernice support this phenomenon, as all attempts made to cast copper in limestone molds, preheated or not, failed (Pernice 1904).

Nonetheless, it appears that the safety and durability issues with respect to the thermal properties of limestone did not stop the ancients from using it as a mold material for casting copper or copper alloys in the form of objects other than ingots, as there have been stone molds recovered from many different regions, including Sardinia, Cyprus,

and Turkey at Troy, all dating to the Late Bronze Age (Craddock et al. 1997: 1). Indeed, Tylecote and others discuss many stone molds from this era used to create jewelry, votive offerings, and flat axes and other tools (Tylecote 1992: 40; Craddock et al. 1997: 1). Tylecote (1992: 39), in a later publication, goes on to say that such molds could have been preheated to help cope with the thermal shock of the rapid introduction of molten metal, and that certain stones would make excellent molds because of their softness and the ease in which they could be carved.

The mountains of Barbagia on the island of Sardinia have also been noted as yielding Bronze-Age stone molds for casting tools and weapons (Balmuth and Tylecote 1976: 195). Clearly, with the discoveries of numerous stone molds for smaller copper or copper alloy objects, and the discovery of the Ras Ibn Hani copper oxhide ingot mold, it appears certain types of limestone was a readily available and viable material for the manufacture of molds used to cast metal objects.

Additionally, excavations at the Late Bronze Age site of Kommos on Crete has turned up large slabs of limestone with circular depressions in the upper surfaces, which have been interpreted as possible bun-ingot molds (Blitzer 1995: 485). Blitzer goes on to say in her report:

“Their resemblance to the type of mold which might be employed in the fashioning of metal bun ingots (both in terms of diameter and depth) is almost too obvious... [The best example] (GS701) occurs in an area of the structure containing probable stone metalworking implements, a clay mold fragment, a number of hearths, and metal debris” (Blitzer 1995: 485).

Further supporting Blitzer’s claims are six copper ingot fragments recovered from the site of Kommos—all from Late Bronze Age deposits (Blitzer 1995: 500), although it is

admittedly strange and unlikely for ancients to have cast copper ingots at Kommos, as there is no evidence of copper smelting facilities at that site.

Upon examining the photographs of these objects in the Kommos report (Shaw and Shaw 1995: 743-44), it is apparent that three of the four supposed bun ingot molds are smoothly rubbed depressions on the surfaces of limestone slabs that would have resulted in too soft a transition from the side of a cast bun ingot to the smooth mold surface. Bun ingots, at least those from the Uluburun and Cape Gelidonya shipwrecks, have much more defined edges than the Kommos molds could have produced. Contrarily, the limestone depressions discussed by Blitzer appear to have been produced as a result of a grinding operation on the limestone slabs. One of the limestone slabs does, however, contain a depression that appears to have resulted from carving with a chisel rather than by abrasion, and is quite different in appearance than the other three supposed bun ingot molds. Contrary to the other slabs, artifact GS 703, with its sharper and more defined shape, seems far more likely to possess the characteristics of a bun ingot mold.

Examination of the limestone oxhide ingot mold at Ras Ibn Hani gives some clues as to how ancient metallurgists may have coped with the apparent instability of limestone when exposed to the heat of molten copper. The limestone used to create the Ras Ibn Hani mold is highly fossiliferous, and it is theorized that the resulting increased porosity in the stone may have helped in dissipating the heat from molten copper, enabling the mold to withstand thermal shock better than other types of limestone (Craddock et al. 1997: 5).

Further supporting the validity of the Ras Ibn Hani mold as a once functioning ingot mold are experiments performed with limestone from the island of Gozo, off Malta, which closely resembles the stone used in the molds from ancient Cyprus and Ras Ibn Hani (Craddock et al. 1997: 4). In these experiments, multiple copper ingots were cast in a mold of Gozo limestone with no pretreatment other than a heating of the mold surface with a blowtorch to remove any moisture that might create a dangerous situation violent volatilization upon exposure to the extreme heat of liquid copper (Craddock et al. 1997: 4). After casting, the only apparent damage the mold sustained was the adherence of approximately 1 mm of the mold surface to the underside of an ingot (Craddock et al. 1997: 5).

The bonding between ingot and mold noted in this experiment suggests that ancient metal workers used substances on the surfaces of the molds in order alleviate the problem of metal sticking to the mold upon solidification. Research shows that in Africa, powdered ash is often spread on the surface of ingot molds to prevent the occurrence of such incidents (Bisson 2000: 103). Interestingly, during excavations at Kition, Karageorghis and Kassianidou (1999: 180) noted large amounts of bone ash in all rooms of the northern workshops and some of the western workshop rooms as well. While no direct correlations between the bone ash and metal production could be made, it does lead to the interesting speculation that perhaps ash was used in ancient times as well as a sort of “non-stick” agent in metal casting.

Evidence for the use of stone molds in the production of copper ingots is not confined to the eastern Mediterranean; finds in Africa’s copper belt also confirm the use

of stone molds for casting copper ingots, albeit much later in the temporal sequence than in the Eastern Mediterranean, and for much smaller ingots. Bisson (2000: 103) states that open molds of heat resistant-stone would be among the materials used to create copper ingots among the tribes of central Africa. Indeed, excavations in Zimbabwe confirm this statement, as they produced a steatite mold used to produce the H-shaped copper ingots (Bisson 2000: 121).

Sand Molds

The scarcity of ancient copper ingot molds surviving to modern times could be easily explained by a simple premise: molds were made from materials that did not survive as molds beyond a single use; specifically, that they were made of sand. Indeed, the presence of over 400 oxhide ingots from the Late Bronze Age and only one mold is most curious. What has happened to the molds used to cast all of these ingots? Every ingot recovered from antiquity has to have a corresponding mold, yet there is only one. How is this possible? One explanation that would allow for the production of a great quantity of ingots and leave no evidence of their creation is sand casting. If copper oxhide ingots were cast in sand, then no direct evidence of their production would have lasted beyond the day of their production.

In Central Africa, the use of sand or earth as a mold material was documented by many observers in the late 19th century. Fred S. Arnot (1889: 238) writes in his travelogue:

“At other mines [copper] is cast in the form of a Maltese cross, the mould being made in the sand by the workers, with their fingers, and out of twenty casts from such moulds scarcely a fourth or an eighth of an inch difference is discernible”.

Bisson, too, notes sand casting among the Yeke, who cast large quantities of copper in ash-lined holes in the ground. He also speaks of the Venda miners of Messina, who cast their bar shaped ingots in the soil by depressing a 1-2 cm thick stick in the ground (Bisson 2000: 98, 102). Even the crude formation of some of the two-handled oxhide ingots from the Uluburun shipwreck has led Pulak to suggest the use of sand as a possible mold material (Pulak 2000: 141).

Although the action is termed ‘sand casting,’ sand molds cannot be made from sand alone, since sand cannot hold a shape very well and molten metal would cause the sand to float, further deforming whatever mold form a metal-caster had fashioned. Instead, green-sand molds are used, which are, in essence, molds that are comprised entirely of damp sand (International Correspondence Schools 1906: 1). Green sand is prepared with sand or silica, to which clay is added as a binding agent (International Correspondence Schools 1906: 6). Put more simply, a sand mold is made of damp sand mixed with clay powder or particles. The function of the clay is to serve as a binding agent that, when water is added, allows the sand to hold its shape.

CHAPTER III

EXPERIMENTATION

Hauptmann et al.'s (2002: 18), analysis of the ingots from the Uluburun shipwreck concluded that bun and oxhide ingots are metallurgically and chemically the same. Indeed, Muhly et al. (1977: 357), twenty-five years prior to the study of the Uluburun ingots, echoed the same sentiment by stating that the difference between bun and oxhide ingots that of was size, not composition. Since it is evident that aside from size there is no discernible difference between oxhide and bun ingots, for practical considerations, the experimentation discussed here will seek to determine mold types used to cast copper ingots in the Late Bronze Age by replicating only bun ingots, rather than the larger oxhide type. Admittedly, it would have been more impressive to cast oxhide ingots, but to do so repeatedly and on the scale needed to test as many mold and pour combinations as possible would have been impractical and economically unwise due to the cost of procuring large quantities of raw copper.

Copper Used

In an effort to ascertain the type of molds Late Bronze Age metallurgists used to produce the copper ingots transported on the Uluburun ship, ingot were cast in various types of molds. Seeking to approximate the ingots from the Uluburun shipwreck as closely as possible, copper from a chalcopyrite ore source was needed. This is the same

ore type from which the raw copper was produced to cast the Uluburun ingots. Obtaining this type of copper from the experiments was imperative, so that some of the impurities found in the chalcopyritic ore remained prior to casting the ingots. The reasoning behind this is that the evolution of gases from the cooling metal would affect the appearance of the ingots in the same as it did in the Late Bronze Age. For this purpose, 23 kg of blister copper shavings were purchased from the Kennecott Utah Copper Corporation and obtained from the Bingham Canyon Mine in Utah, which produces copper from a chalcopyrite ore. Kennecott shipped shavings from fire refined anode copper ingots, which are then used to obtain 100% pure electrolytic copper through electrolysis. The shavings are roughly 99% pure copper, with trace amounts of lead (0.05-0.30%) and arsenic (0.02-0.11%), based on specifications provided by the Kennecott Utah Copper Corporation.

Furnace and Fuel

To melt copper for casting, a furnace capable of generating the heat required to reach the melting point of copper was necessary. Since the goal of this experiment is not aimed at replicating the smelting technologies from the Late Bronze Age, but merely testing copper ingots cast in molds of various materials, it was not necessary to build a replica of an ancient furnace. Instead, the simply designed and easily constructible furnace described in “The Flowerpot Crucible Furnace” (Oliver 2002) was chosen for the project. The furnace design consists of a 12-inch terracotta flowerpot sunk into a

five-gallon tin filled with cement. In the bottom of the can a hole is drilled through the cement and into the terracotta pot to provide access for blast air. The lid of the furnace consists of sheet metal riveted onto the original metal lid, allowing for about three inches of cement to be poured in, which provides a much sturdier and better insulated top for the furnace. A hole was left cleared in the center of the lid to act as an exhaust vent. Blast air was fed into the furnace from an electric hairdryer through a PVC pipe and was distributed through the bottom of the furnace with the aid of angle iron drilled with holes.

However, in order to generate the heat to melt the quantity of copper needed for the casting experiments, the design of the furnace discussed above had to be modified (Figures 1 and 2). The basic concept for the furnace remained the same, however, roughly three quarters of a 55-gallon drum was used instead of a five-gallon tin, and a much larger terracotta pot was sunk in place. To deliver the blast air to the combustion chamber, a capped iron pipe with many holes drilled along its length was used in place of the PVC pipe. There was one attempt to use the PVC pipe, but it quickly melted under the extreme heat of the newly designed furnace. Blast air was produced by a blower from a salvaged furnace that was fitted with a rheostat to control the amount of air being forced into the furnace.



FIGURE 1. The scratch-built furnace used for these experiments (photo by author, 2006).



FIGURE 2. Furnace with crucible prior to removal of molten copper (photo by author, 2006).

A number-10 graphite crucible (40 cm height, 33 cm top diameter, 23.2 cm bottom diameter) was used to contain the copper during melting. This crucible, which holds 10 pounds of aluminum, has sufficient volume to hold more copper than is necessary to cast any of the bun ingots from the Uluburun shipwreck.

The fuel choice for the furnace was a difficult decision. “The Flowerpot Crucible Furnace” (Oliver 2002: 15) calls for the use of regular charcoal to fuel the burn, however, this design is primarily aimed at melting aluminum, which has a far lower melting point than that of copper. Nonetheless, in the initial trials the furnace was fired with charcoal. As expected, the furnace had no problem melting aluminum in a matter of minutes. However, when attempting to melt copper, difficulties arose. The furnace was indeed capable of reaching the melting point of copper when fueled with nothing other than charcoal, but in reaching that temperature, the fuel was consumed quickly and the copper resolidified prior to melting completely. To remedy this problem, two solutions were available: build a bigger furnace capable of holding more fuel, or use a hotter and longer burning fuel. Since the design of the furnace had already been augmented, and increasing its size further would prevent it from being portable in nature, a hotter and longer burning fuel was sought. For this, bituminous coal was chosen, as it is a good compromise between charcoal and harder, dirtier burning coals. Charcoal was not completely eliminated from the design, as it was used in the bottom of the furnace to ignite the bituminous coal. Once the charcoal and bottom coals began burning, the blower was then powered up and the furnace became fully operational.

Molds

Literature research presented earlier details the materials possibly used to manufacture the molds in which the Uluburun ingots were cast. Based on that research, molds made of clay, sand, and limestone were chosen for experimenting. All of the molds were made to roughly approximate the size of a typical bun ingot from the cargo of the Uluburun shipwreck. In more precise terms, the manufactured molds varied from 16-21 cm in diameter and were at least 5 cm deep in order to accommodate sufficient copper to replicate bun ingot at full scale.

To create the clay mold (Figure 3), a substantial amount of chopped straw, crushed charcoal, and sand was added to standard gray potter's clay. The choice for these materials was based on Freestone's (1989:157-160) article concerning refractory materials in antiquity; specifically the portions concerning clay molds and crucible. The inclusions were mixed into the clay so as to evenly distribute them throughout the clay matrix. The mold was then allowed to air dry until hard before being baked for a day over an open gas stove. The resulting mold basin (portion in which the copper is to be poured) has a maximum diameter of 22 cm, while the bottom of the basin has a diameter of 14 cm and a depth of 6 cm.



FIGURE 3. Clay mold after the casting of the first ingot (photo by author, 2006).

The limestone (Figures 4 and 5) mold was carved from a 93-pound slab of Austin limestone, measuring 41 cm long, 31 cm wide, and 14 cm. high. The mold basin has a top diameter of 21 cm, a bottom diameter of 18 cm, and a depth of 6 cm. The mold basin was carved with the aid of a pneumatic chisel from Texas A&M University's Conservation Research Laboratory, Riverside Campus, College Station, Texas. All the casting work was carried out at this facility. The sides of the basin are quite smooth, while the flat bottom of the cavity is noticeably rippled from the tip of the chisel. This rippling, which should be evident in the mold surface of the copper ingots when cast, was also noted on the Ras Ibn Hani mold is Syria (Lagarce et al. 1983: 277).

Green sand was used to form the sand mold (Figure 6). For this experiment, a mixture of roughly 7:1 sand to bentonite clay dust was used, with the addition of 5% water by weight. Large, coarse sand was chosen for this purpose, since it would more readily allow the escape of gases from molten copper than fine sand. The use of fine sand, since it is more efficient at trapping gases, could cause the casting to blow (International Correspondence Schools 1906: 6). Bentonite clay powder was obtained from grinding cat litter (Pet Gold Cat Litter brand) in a coffee grinder. Many cat litters, including the brand used here, are made from small chunks of bentonite clay that readily coagulate around wet urine and moist feces from cats. This is exactly the property needed to bond sand with added moisture from water. The cat litter was ground to a size that matched the coarseness of the sand to facilitate mixing.



FIGURE 4. The author carving the limestone mold with a pneumatic chisel (photo by John Swanson, 2006).



FIGURE 5. Limestone mold, split from thermal shock during the casting of the copper ingot (photo by author, 2006).



FIGURE 6. Green sand mold prior to the pouring of molten copper (photo by author, 2006).

CHAPTER IV

CASTING EXPERIMENTAL INGOTS

Casting of the Ingots

Pouring liquid copper, which melts at 1083 °C (1981 °F), into molds of green sand, clay, and limestone resulted in specific reactions as the copper encountered the different mold materials. For all castings, the copper used was heated above its melting point. To achieve this, the copper was left in the ignited and running furnace for five minutes after it transformed into a total liquid state. The crucible containing the copper was then removed from the furnace and the copper poured into a molds (Figure 7) by varying the pouring procedures described in the previous chapter.

The most agitated reaction between copper and mold came from casting in limestone. When molten metal was poured into the mold, crackling and popping sounds were generated and the metal bubbled profusely as long as it remained in a liquid state. The high rate of bubbling and instability of the reaction was likely due to a combination of factors; including the breaking down or decalcining of the limestone mold itself, and the soluble gases within the copper coming out of solution and escaping the molten copper as it cooled. Limestone becomes thermally unstable at temperatures above 900 °C as calcium carbonate decalcines into lime, which results in the evolution of carbon dioxide gas. The reaction increases in volatility as temperatures increase beyond 1000 °C (Craddock et al. 1997: 1).



FIGURE 7. Pouring molten copper from the crucible into a sand mold (photo by John Swanson, 2006).

Despite the relatively violent reaction between molten copper and limestone, casting in this material was not dangerous, as the reaction did not spew molten copper droplets beyond the confines of the mold or of the sand box in which the mold sat. As it cooled in the limestone, the copper remained liquid for at least one minute, continuing to bubble as long as it remained in a liquid state.

Pouring molten copper into a clay mold provided insight as to how sudden exposure to extremely high temperatures can affect a mold made of clay. As soon as the copper came into contact with the mold, an audible sizzling sound was noticeable. When sufficient copper had been poured so that the bottom of the mold was covered with molten metal, the violence of the reaction between liquid copper and clay became even more apparent; the copper bubbled and spewed tiny bits of molten metal. The result of casting in this material was second only to casting in limestone with respect to the violence of the reaction. Amidst the turmoil of agitated molten copper, thin flakes of clay appeared and floated on the surface, an apparent sign that the surface of the mold was breaking down from exposure to intense heat.

Casting in green sand resulted in the least significant reaction between mold and molten copper. The relative non-violent reaction of casting in green sand came as a surprise, since it was the only mold to contain moisture. Water, when exposed to extreme heat, will instantly vaporize. This caused concern that the interaction between molten copper and water might potentially be dangerous to those nearby when the copper ingot was cast. These fears, however, proved to be unfounded as the copper settled smoothly into the mold with very little bubbling. The only observable agitation

occurred at the moment when the copper was poured into the mold. Even this reaction, however, was mostly due to the sloshing effects of the liquid metal as it fell from the crucible into the mold basin. Generally speaking, the copper cooled calmly, as evidenced by the smooth appearance of the exposed or “rough” surface on the sand-cast ingots.

Single Pour Ingots

Sand Mold, Single Pour

Macroscopic Observations

The term single pour ingot defines an ingot cast using one single, continuous pour of molten copper from the crucible. The purpose of creating ingots from a single pour is to establish a baseline to which both single and multiple-pour ingots can be compared to the Uluburun collection.

A circular depression of 17.7 cm in diameter was made in a green-sand mixture using a cooking pot. This depression served as the mold form or template for casting a copper ingot in green sand. The resulting ingot has a uniform diameter of 17.7 cm, a maximum thickness of 3 cm, a minimum thickness of 1.4 cm, and weighs 4.26 kg.

The upper exposed, or rough, surface of the ingot exhibits slight blistering. Generally, the rough surface of this ingot is smooth when compared to the ingots from the Uluburun shipwreck. The rough surface of this sand-cast ingot appears mostly to consist of a thin copper crust, as evidenced by the many blows on it and its fragile nature. There is a low, but wide, shrink depression along the edge of the thinnest portion

of the sand-cast ingot. Along the edge of the thicker portion of the ingot is a thin line of rough copper, which could also be construed as a cooling ridge.

The mold surface has very few, if any, gas porosities. There are two prominent swells that were either caused by the molten copper displacing loose sand in the mold basin or from preexisting imperfections in the mold. The evidence of a mold ridge around the edge of the entire mold surface of the ingot can be seen. Adhering to the mold surface is a thin layer of bentonite clay powder from the sand mold; but, there does not appear to be any sand attached to the mold surface. Near the edges of the mold surface there are several small slag inclusions, which appear to be the only visual impurities in the ingot.

The lack of porousness of this ingot is due to the material in which it was cast. Of the three mold types used in this study, a sand mold would allow trapped gases to escape more freely than any of the other mold types. This is because sand molds consist of thousands of tiny grains of sand, each with space between them to allow for the passage of gases evolved from the molten copper. The other two mold materials, however, are made of a less porous composition and are therefore much denser. Therefore, molds of stone and clay would more effectively hinder the escape of gases evolving from the cooling ingots.

Sectioning the ingot to reveal its inner structure confirmed what had been suspected about its appearance, based on the lack of porosity observed on both of its surfaces. The section revealed an inner structure nearly devoid of any gas pores that could be detected by naked eye. That is not to say that microscopic examination would

not reveal small gas pores, but, visually, it appears that the only trapped gases seen were located just below the crust of the rough surface. Casting an ingot in a single pour in a sand mold, therefore, results in an ingot devoid of porosity throughout its body, with the exception of a few small cavities created by trapped gases just beneath the crust that forms atop the cooling ingot. This thin, outermost layer, with little gas pressure from beneath, seems to have been sufficiently strong to block the escape of what few gases were trapped below it at the time of its formation.

Microscopic Observations

An examination of this ingot specimen under a microscope supports initial observations made concerning its dense appearance. Indeed, microscopic analysis confirms that the body of this ingot contains little gas porosity, except just below the rough surface. The porosity near the rough surface is not so much imbedded in the copper matrix, but resembles gaps left in the copper as a result of escaping gases. These pockets of trapped gas are approximately 30 μm in size, which is near the small end of the size scale with respect to copper grains formed in this sand-cast ingot. The occasional pores observed outside the vicinity of the rough surface were small, no larger than 10 μm across. In one instance (Figure 8), the pores were arranged in a vertical

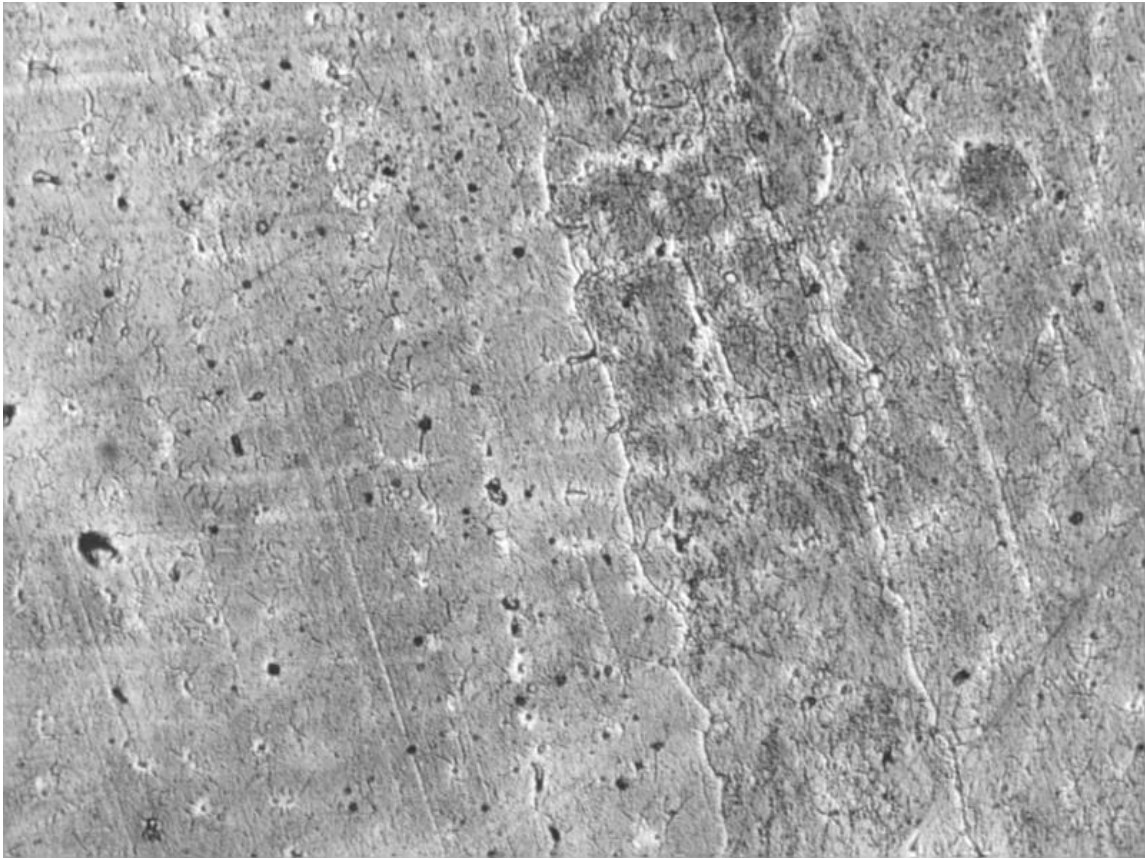


FIGURE 8. Directionality within the sand mold, single-pour ingot at 200x magnification. Note the alignment with which the copper grains are configured (photo by author, 2007).

fashion, indicating the upward flow of gases seeking escape from the molten copper solution.

Copper grains are not equiaxed (Scott 1991: 8) in this sample, varying substantially in size and shape. Grain size throughout the ingot varies from 80 μm to 30 μm . This size discrepancy does not just occur between the grains near the rough surface and those deeper in the matrix, but can be seen throughout the body of the ingot.

There is significant directionality within the crystalline structure near the center of the ingot (Figure 8), indicating more rapid cooling than was noted in Hauptmann (2002) et al.'s study of the Uluburun ingots.

Aside from the low frequency of pores throughout the ingot, magnification reveals that small portions of the mold material were likely imbedded in the copper matrix as well. Certain small (less than 5 μm) gray colored inclusions can be seen in areas of the ingot with slightly increased porosity (Figure 9).

Given the apparent color of these inclusions, it is likely that they may consist of small particles of bentonite clay powder that was used to bind the sand and facilitate the forming of the mold. Aside from their gray appearance, these particles stand out because they are lighter in color than their background and are encircled by a small black corona. Unlike the small inclusions noted in the single-pour ingot cast in a limestone mold, the coronas surrounding these inclusions are sharp, not blurred.

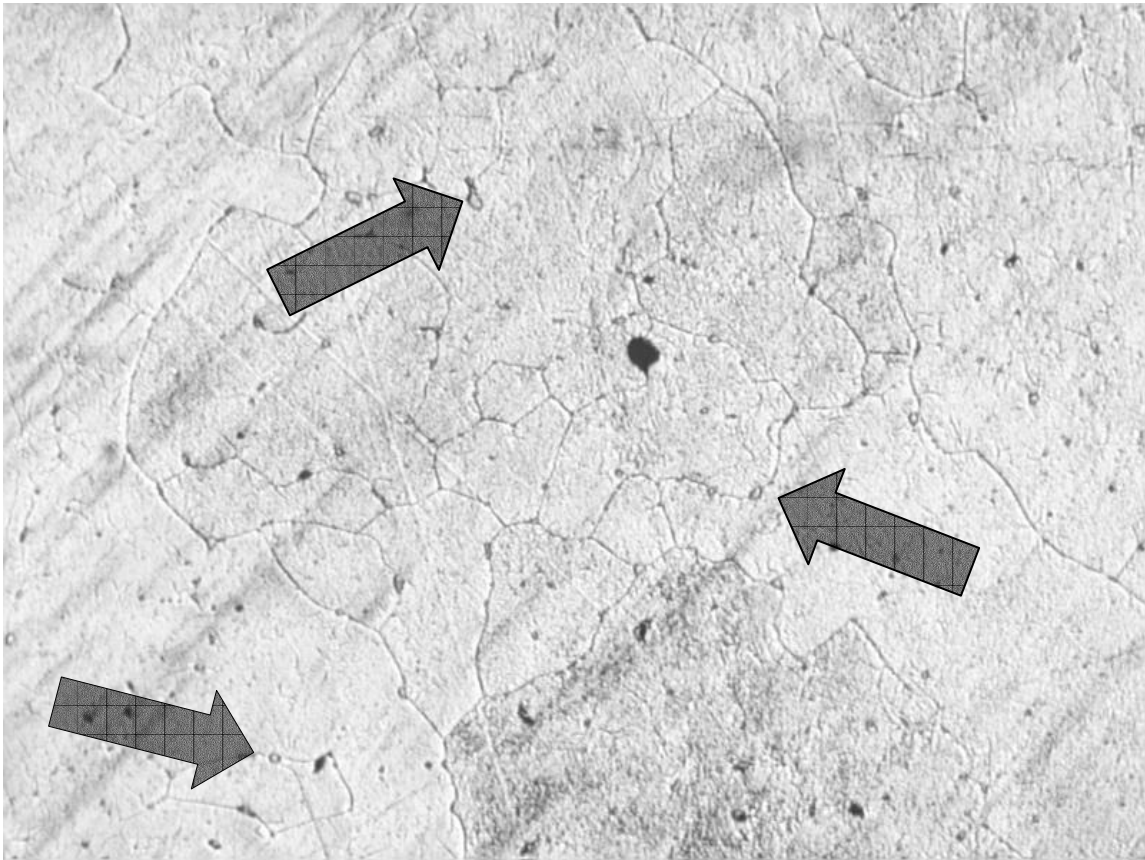


FIGURE 9. Sand mold, single-pour ingot at 200x magnification. Note the numerous gray-colored round inclusions (photo by author, 2007).

In addition to the relative denseness and lack of porosity in this ingot, there do not appear to be many precipitates along the grain boundaries. Instead, the majority of what few precipitates are observed can be seen within the boundaries of the copper grains. Lack of precipitates along the grain boundaries and, instead, within the grains, suggests that this ingot cooled more rapidly than those from the Uluburun shipwreck; so rapidly that what precipitates did form were not able to migrate beyond the grain boundaries before the copper solidified. Indeed, after examining this ingot under magnification and with the unaided eye, it is apparent that there are some significant differences between it and those ingots analyzed from the Uluburun shipwreck.

Limestone Mold, Single Pour

Macroscopic Observations

The single-pour ingot produced by casting in the limestone mold had an irregular shape, as the mold was carved by hand, without the use of a pattern. The maximum diameter of this ingot is 18.7 cm, and the minimum diameter is 16.4 cm; its thickness is varied between 3.4 cm and 2 cm. The cast ingot weighs 4.40 kg.

The limestone mold in which the ingot was cast suffered a crack that separated it into two halves. The crack was formed in an early phase of the experiment when two ingots were cast in the mold that were too thin to be considered accurate representations of the Uluburun shipwreck ingots. An attempt was made to join the two halves of the limestone mold so that the crack would not be a factor during additional casting.

However, this attempt proved to be somewhat futile, as evidenced by a large fin resulting from molten copper seeping into the crack that separated the two halves of the mold. Nevertheless, as unsightly as this casting defect is, it should not affect the analysis and results of the casting experiments. In fact, this anomaly could be fortuitous and result in information leading to the identification of similar fins on the mold surfaces of ingots from antiquity.

The rough surface of the cast ingot contains many low blisters accompanied by small blows. There are several visible large slag and coal inclusions on the rough surface, including one over three centimeters across near the center of the ingot. While the inclusions likely resulted from sloppy casting (intense heat caused the lid to break down and slough pieces of the sand/cement mixture into crucible), the large blisters and unusual protuberances on the ingot's rough surface probably resulted from the interaction of the molten copper with the limestone mold. A shrink-depression is present on the ingot in the form a smooth, flat ring around the edge of the rough surface. The shrink-depression is distinct and not marred by blisters and blows seen on the rest of the rough surface. The depression likely resulted from the ingot cooling more rapidly around its perimeter due to being in contact with the mold, and thus preventing the escape of gases from this cooled region on the surface.

The rough surface also contains two distinct protuberances near the edge of the ingot. These two protuberances are not easily explainable, as they do not appear to be the result of cooler copper spilling from the crucible to an already hardened surface. Since

they are near the faster cooling edges of the ingot, it is possible that they resulted from agitated copper freezing in odd shapes before settling smoothly into the copper matrix.

Immediately noticeable on the mold surface of the limestone-cast ingot are inclusions of white, powdered limestone bits from the mold. The limestone powder is concentrated around areas of the mold surface marred by gas porosity. The juxtaposition of these two features indicates that the porosity is perhaps more the result of the breakdown of the limestone and the resulting evolution of carbon dioxide, than soluble gases trying to escape the cooling copper. The porosity visible on the mold surface occupies the central portion of the ingot, leaving the edges to appear solid. The obvious explanation is that the copper near the edges of the mold cooled more quickly, as it had traveled farthest over the cool mold, and no new molten copper was poured on top of it to maintain its molten state. Since the edges of the ingot cooled faster than the center, they did not maintain the elevated temperatures necessary long enough to cause the limestone to break down and produce the carbon dioxide gas that is likely to have caused the porosity on the mold surface. This observation is confirmed by the limestone mold as well, since it can clearly be seen that the majority of the surface degradation is near the center of the mold, leaving the edges relatively intact.

The most prominent feature of the mold surface of the limestone-cast ingot is the fin bisecting the ingot's surface. This fin resulted from the limestone mold when it cracked into two halves by previous casting attempts that yielded ingots of less than desired thickness. While this feature is not something that will prove diagnostic to identifying all ingots cast in limestone, it does provide comparative material when

examining archaeological ingots suspected of bearing similar casting defects. The fin is not a solid formation, but resembles a splatter tacked to the mold surface, with smooth, rounded, globular tendrils reaching out at odd angles.

Sectioning the ingot revealed an interior substantially more dense than what might be suspected based on the extant porosity noted on both the rough and mold surfaces. Indeed, the interior of the ingot is almost completely devoid of evidence of trapped gases, except at very near the rough surface, where there are a few, tiny, gas pores. This is an indication that limestone molds might be more efficient than anticipated in allowing the dissipation through the mold material of gases evolved during the cooling of an ingot, and not just released through the exposed surface of an ingot.

Although the rough surface of the ingot does not contain flakes of limestone similar to those seen on the mold surface, sectioning the ingot revealed the instability and frailness of limestone as a mold material by exposing a small fleck of the mold trapped within the copper matrix. Located near the rough surface, this trapped fragment of the mold within the ingot indicates the mold began to break down early in the casting process, allowing an inclusion of limestone to rise nearly all the way to the rough surface. With the strong presence of limestone adhering to the mold surface of this ingot, it is actually surprising to discover only one small fragment of limestone imbedded in the interior of the ingot. Perhaps this is an indication that the limestone does not flake off as it breaks down, much in the way of a clay mold, but rather dissolves into a powder that is destroyed as it mixes with the copper.

Microscopic Observations

Microscopic examination of the ingot confirms many of the observations made during its macroscopic analysis. When viewed under 200 magnification, the sample from the ingot revealed a specimen relatively void of porosity. What few pores were detected, were not excessively large. In fact, they did not seem to exceed the average size of the copper grains.

Grain size was constant throughout the corpus of the ingot, varying between 60 to 140 μm , with an average size of 80 μm . The only exception to this observation was very near the rough surface of the ingot, where cooling would have occurred much faster due to the direct exposure to the atmosphere. There, as would be expected, grain size was noticeably smaller than that of the rest of the ingot, where cooling was more gradual. Under the microscope, the grains appear to have regular size and shape, and can be described as equiaxed.

Near the bottom of the ingot, small inclusions of what appear to be tiny portions of charred limestone or the ash used to line the mold basin were detected (Figure 10). These inclusions are small (less than 10 μm in diameter), circular, and surrounded by a blurred, seemingly charred corona. They are dissimilar from those found in the single-pour ingot cast in sand, since their coronas are fuzzy, not sharp and fine as with those observed in the single-pour, sand-cast ingot. These inclusions, however, are unique to this sample and oddly do not appear in any of the other two iterations of ingots cast in a limestone mold.

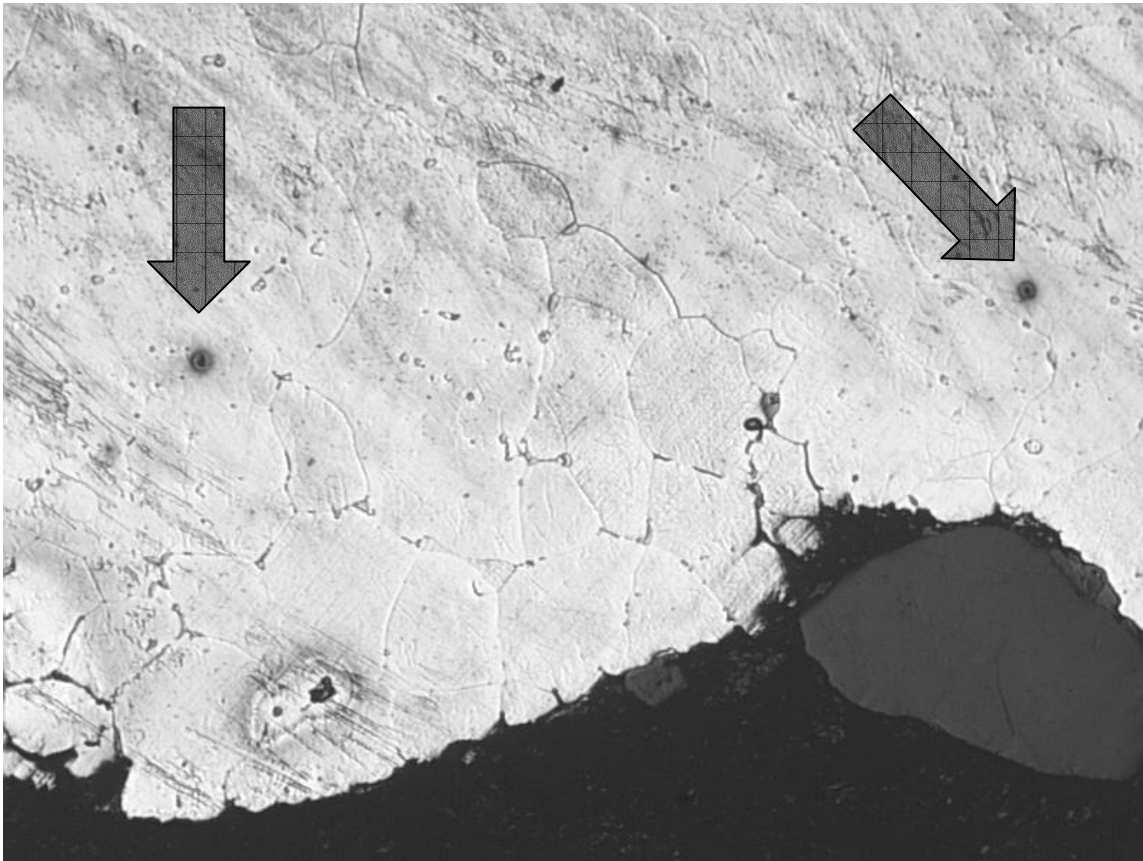


FIGURE 10. Limestone mold, single-pour ingot at 200x magnifications. Notice the fuzzy, dark halo surrounding two inclusions (photo by author, 2007).

There do not appear to be a significant amount of precipitates within the copper matrix of this ingot. In fact, when compared to photos taken of the samples taken from the Uluburun shipwreck ingots, there are relatively few precipitates in this ingot, especially along the grain boundaries. Not unlike the single-pour ingot cast in sand, a small number of precipitates within the borders of the individual copper grains are evident.

Clay Mold, Single Pour #1

Macroscopic Observations

The first ingot cast in a clay mold has a different general shape than those cast in limestone and sand. It weighs 3.72 kg, has a maximum diameter of 18.1 cm, a minimum diameter of 16.8 cm, a maximum thickness of 3.8 cm, and a minimum thickness of 2.9 cm. While the limestone and sand ingots have nearly vertical sides, the clay-cast ingot is lens shaped, with no discernible sides, since the mold surface gradually tapers into the rough surface. This shape is likely the cause for the most prominent feature observed on the rough surface. The center of the rough surface rises higher than the edges, somewhat resembling a low hill rising from flat a plain. This swell is likely the result of the unique profile of this ingot. The lens-shaped section makes the edges significantly thinner than the center of the ingot, resulting in the cooling and solidifying of the edges at a much faster rate. In the instance of this ingot, the edges cooled before all the molten metal was poured from the crucible. Given that the edges had cooled and hardened before all the metal could be transferred, additional molten copper could not be absorbed by the liquid

metal in the mold as readily in the case of the other ingots. Thus, the molten copper began to pile up in the center, forming a swell in the center of the cooled and solidified ingot.

It has already been postulated the shape of the clay-cast ingot resulted in the periphery of the ingot cooling and hardening much faster than its central region. The rapid cooling near the edges caused the copper to solidify faster and more densely than the center portion, and therefore the edges are flatter and do not contain as many blisters as the core of the rough surface. There appear to be several inclusions of semi-molten metal near the center of the rough surface that are incorporated into the copper matrix. The central swell on the rough surface is made more prominent by blistering, which possibly resulted from the effervescence of sulfur dioxide gases in the molten copper as the metal cooled. The blisters on the rough surface are accompanied by many small blows, which were no doubt caused by the evolution of gases.

The mold surface exhibits multiple traits that are unique to ingots cast in clay molds. Due to the extreme heat of the molten copper when it came into contact with the surface of the clay mold, many sections of the mold flaked off or spalled as the copper was poured. Evidence for this is present not only on the mold, but on the mold surface of the ingot as well. On one corner of the clay-cast ingot (Figure 11) flakes of the mold became incorporated into the mold surface as the molten copper flowed around them and hardened. When examining an ingot freshly cast in a clay mold, such a feature would be self-evident, as the clay flakes would still remain attached to the mold surface. However, millennia of erosion, either on the bottom of the sea—as is the case with the Uluburun

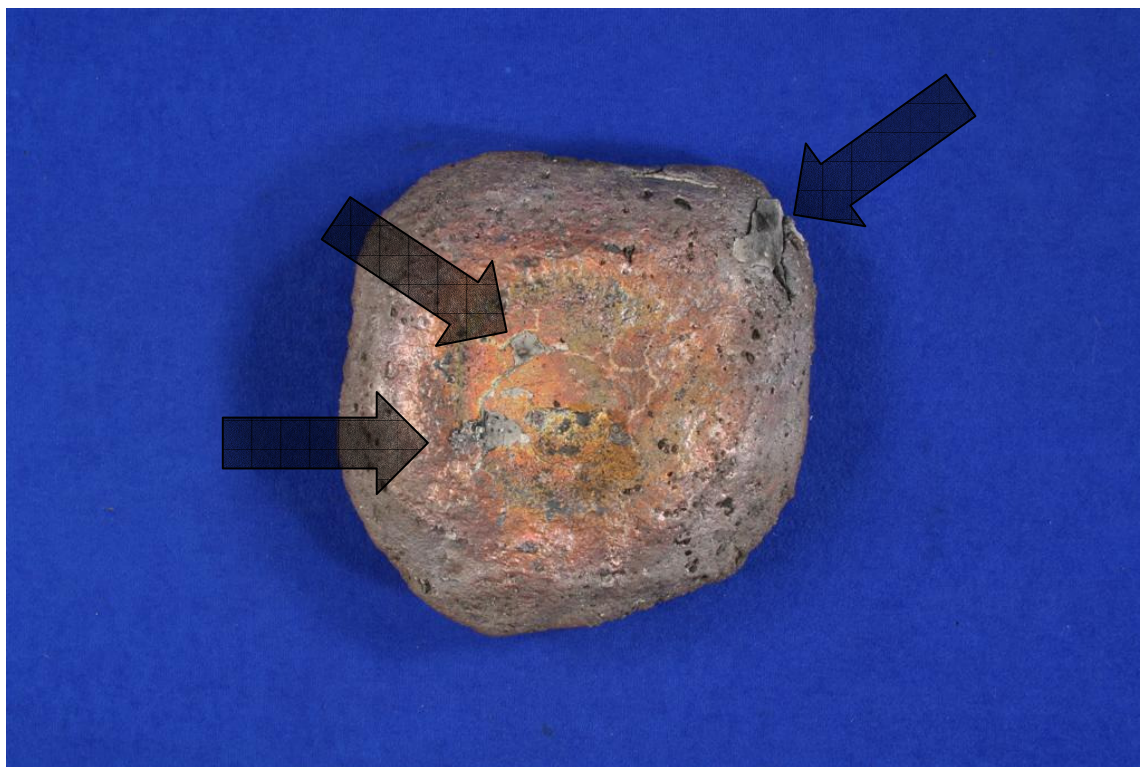


FIGURE 11. Fragments of clay from mold embedded in the center and upper right of the ingot
(photo by author, 2006).

ingots—or in the soils of the lands bordering the eastern Mediterranean and elsewhere, it is not unlikely that small flakes of clay, which once adhered to an ingot, would have disappeared. Despite the absence of mold remnants on archaeological ingots, those cast in clay molds should display evidence of certain features, such as the cavities created by dissolved clay fragments where copper froze around pieces of clay.

More evidence of the clay mold flaking and spalling is seen near the center of the ingot. The nature of clay as a mold material creates jagged, sharp breaks in the surface of the mold when it breaks. Even if no clay adheres to an ingot, evidence of these sharp breaks may be observed as impressions on the mold surface. The mold surface of the clay-cast ingot contains many thin, slightly raised, vein-like ridges where liquid copper seeped into cracks formed in the mold during casting and then hardened. These ridges seem to be unique to the ingot cast in clay and were not observed on ingots cast in sand or limestone.

Since the clay mold was shaped by hand without the use of forms or tools, the edges of the casting basin retain many finger marks left while forming the mold when it was still damp. Impressions of these marks can be seen and felt on the surface of the clay-cast ingot corresponding to those on the mold (Figure 12). Several circular plano-convex discoid from the Uluburun shipwreck (KWs 1122, 2364, 2568, 2792, 3698, and 4409), all identified by Pulak as mold siblings exhibit similar features that have also been interpreted as finger mark impressions transferred from the clay molds to the ingots during casting.



FIGURE 12. Finger impressions on the clay cast ingot (edge, left side) imparted from the clay mold (photo by author, 2006).

The clay-cast ingot displays relatively little, if any, gas porosity on the mold surface. What little porosity there is can only be seen near the perimeter of the ingot. There are, however, many additional holes near the perimeter of the mold surface that should not be construed as gas pores, since they appear to be filled with remnants of spent fuel from the furnace and coal ash used to line the mold to prevent the copper from sticking to the clay mold.

It should also be noted that the copper on the mold surface of the clay-cast ingot has a distinctly more red or copper-like appearance at the center where the ingot is thickest and, therefore, would have remained hotter and molten longer. This two-toned coloration of the mold surface is another unique feature that is not present on the ingots cast in sand and limestone and is unique to the clay-cast ingot. It should be noted, however, that this coloration may not necessarily be a function of the mold material, but as a result of the unique lens shape of the ingot where the center is significantly thicker than the edges. It may even be argued that the color change in the mold surface marks the boundary between the thick and thin portions of the ingot.

Given the lack of porosity on the surfaces of this ingot, it was surprising to see the high frequency of trapped gases in the ingot when sectioned. Cutting off a portion of the side revealed an ingot with extensive inner porosity. Gasses appear to have been trapped throughout the thickness of the ingot and along the edges of all surfaces.

Equally interesting as the extant porosity, is the variation in which it occurs. Most notable is a large cavity beneath the rough surface. Of all nine ingots cast in these experiments, this ingot retains the largest hollow resulting from trapped gas. Not

surprisingly, the pocket of trapped gas resulted in a corresponding swell on the rough surface of the ingot. Observing the correlation between a large gas pore and a corresponding surface swell leads to the speculation that swells on the rough surfaces of other ingots suggest the presence of large gas pores beneath the surface. If this assumption is correct, then even larger cavities exist in other ingots, although no sections were cut through swells in order to confirm this postulation.

Perhaps more intriguing than the singular large cavity near the rough surface is the presence of many small to medium-sized pores just beneath the mold surface of the ingot. The appearance of these pores suggest the presence of effervescing gases immediately following the pouring of the molten copper into the mold. The gases were likely trapped by the weight of the copper and were blocked from escaping through the mold due to lack of porosity of clay. Therefore, the gasses remained trapped near the edges of the mold surface since they could not travel up through the copper or out through the clay mold.

The presence of many small pores throughout the thickness of the ingot imply a similar situation, with respect to their formation, to that of the pores lining the edges,. These pores are likely the result of soluble gases evolved from the middle portions of the ingot. The slightly smaller surface area of this ingot, compared with that of the second ingot cast in a clay mold, could result in the faster formation of a gas-trapped surface crust. The faster surface cooling time, coupled with the added central thickness of the ingot, could conceivably create a more favorable environment for trapping gases. This is because gases in the copper would have farther to travel if seeking escape through the

open surface of the mold, but less time to do so, as a crust of metallic copper would have formed sooner and trapped the gases. This combination of factors is probably the main contributor to the high porosity of this ingot.

Microscopic Observations

Grain size on this, the first of two single-pour ingots cast in a clay mold, is slightly smaller than the two previously discussed ingots cast in limestone and sand. Grain size varies from 30 to 80 μm , with an average sized grain roughly 60 μm in diameter. The grain size near the rough surface is smaller than that of the other ingots discussed thus far, averaging 35 μm . The cause for the reduced grain size is predictable, due to the faster rate of cooling of copper exposed to air. Despite the decreased size of the copper grains, they are nonetheless equiaxed and do not differ in shape or size within the ingot.

Porosity in this ingot is significantly greater than that of either ingot cast in limestone or sand. Not only is the number of pores increased, but the size of the pores are also greatly enhanced. Many pores were noted with diameter's in excess of 500 μm . In addition to the large pores, the number and frequency of precipitates is also higher and occur primarily along the grain boundaries, as opposed to within the individual copper grains. With respect to porosity and precipitates, the ingot appears to be a close approximation to those recovered from the Uluburun shipwreck.

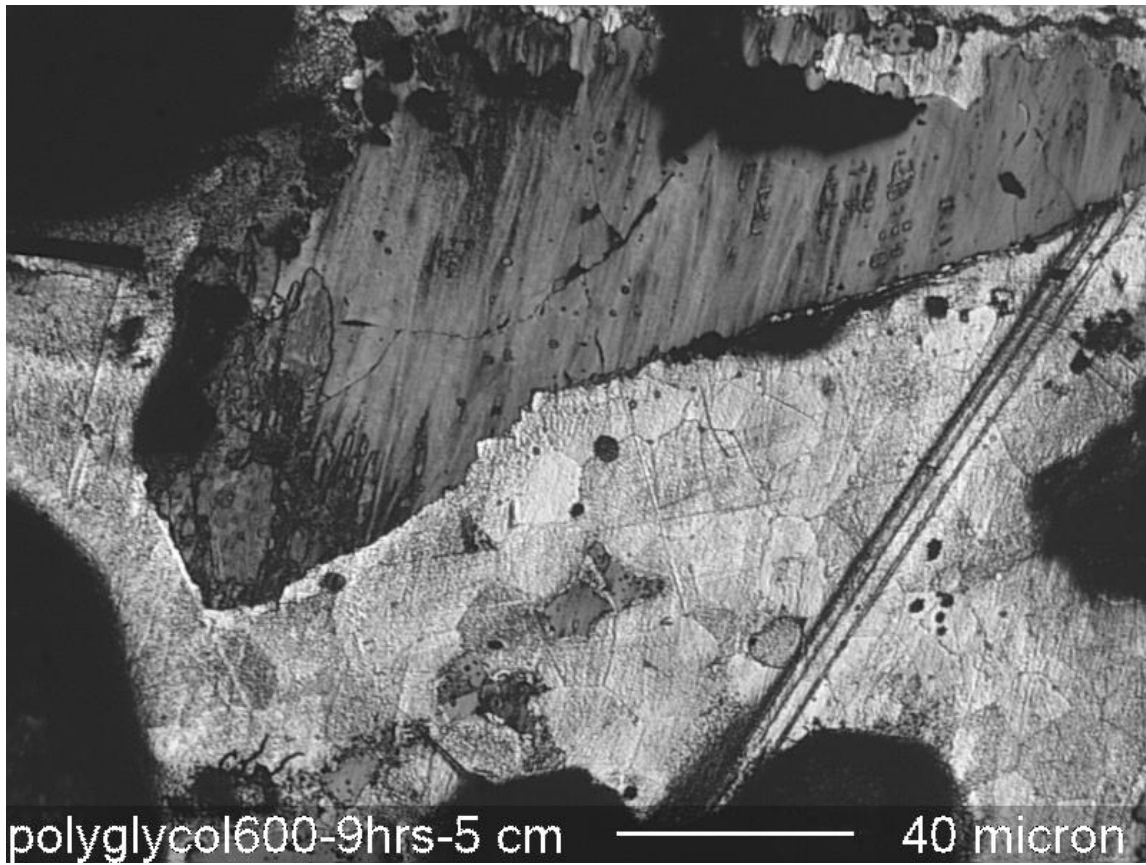


FIGURE 13. Magnified (200x) view of clay inclusion in the first single-pour ingot cast in the clay mold. Scale not accurate (photo by author, 2007).

Aside from the increased porosity of the ingot, there are many inclusions to be noted as well. Most notably, there is a large piece of clay from the mold imbedded in the copper matrix near the rough surface (Figure 13). The inclusion of such a large piece of mold material itself was something that proved to be rare for these experiments, and may prove to be a most diagnostic feature if ever discovered in ancient ingot specimens. The clay inclusion is made evident from the surrounding copper by its darker coloring, distinct borders, and its display many scuff marks from polishing during the sample's preparation.

As with the two previously discussed ingots, there again appears to be smaller inclusions, this time of an unknown material, surrounded by a darkly colored corona (Figure 14). The inclusions and coronas vary slightly in magnitude, but maintain an average size of roughly 10 μm in diameter. There does not appear to be a discernible pattern to the locations of the inclusions within the sectioned sample.

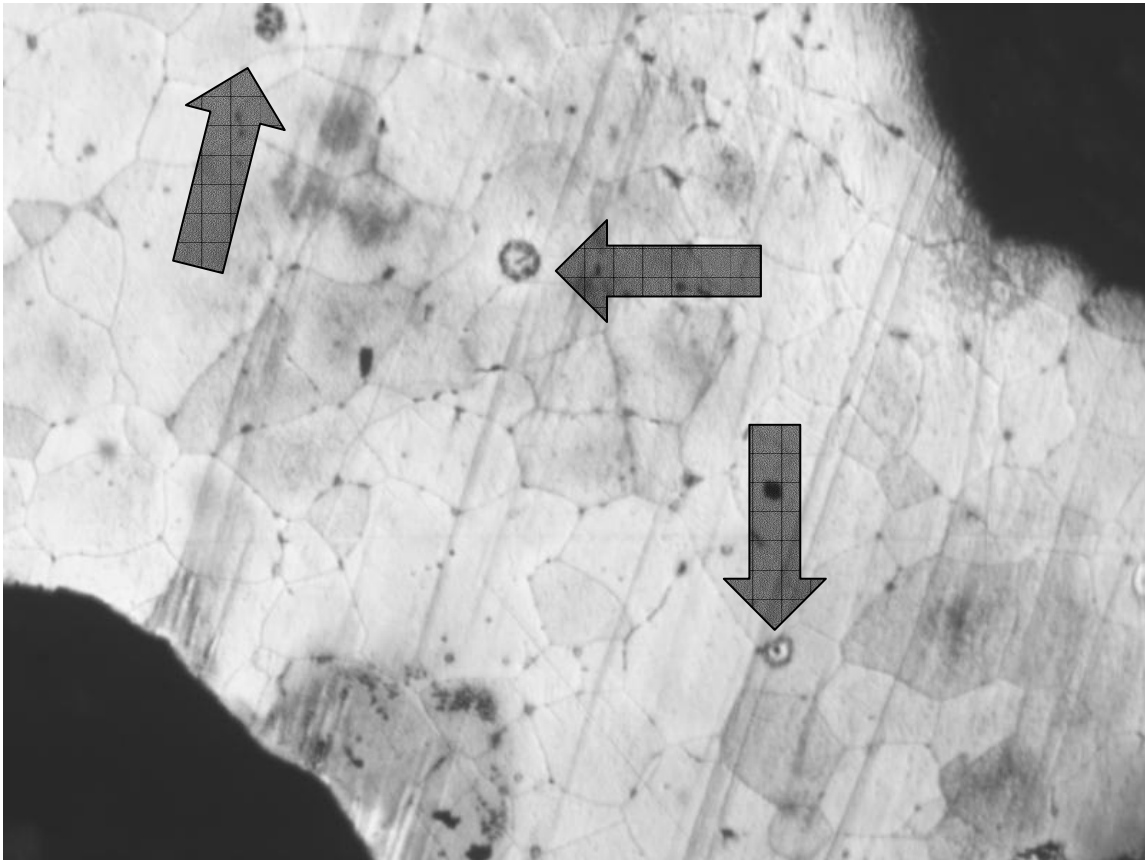


FIGURE 14. Clay mold, single-pour at 200x magnification. Three inclusions surrounded by a dark, halo-like corona in the copper matrix (photo by author, 2007).

Clay Mold, Single Pour #2

Macroscopic Observations

Having already cast one ingot in a clay mold, the purpose of casting another was to more closely replicate the shape of the sand and limestone-cast ingots, so they can be compared more meaningfully. The second clay mold ingot has a maximum diameter of 18.2 cm, a minimum diameter of 17.4 cm, a maximum thickness of 3.2 cm, a minimum thickness of 2.8 cm, and weighs 4.0 kg. Since the first ingot cast in a clay mold was lens shaped, the mold used to cast the second ingot in clay was constructed to have a flatter bottom and steeper sides, thus rendering ingots similar in shape to those cast in limestone and sand mold. Moreover, since the previous ingot cast in clay was one of the first ingots cast in these casting trials, a second casting in clay would provide a chance to better observe how molten copper reacts with a clay mold.

The rough surface of the second clay-cast ingot contains many inclusions of thin clay flakes spalled from the mold basin. In addition to the clay flakes, portions of the perimeter of the rough surface are covered with a thin crust of metal, concealing pockets of air. Nearer the center, the surface thickens and several large swells are present, likely resulting from the pressures of soluble gases that pushed upwards seeking escape into the atmosphere. The coarse nature and extensive blistering of the rough surface is almost certainly a function of the inability of the mold material to let gases escape anywhere but through the rough surface while the copper was still molten. As soon as a thin crust

developed on the rough surface, the force of the trapped gases beneath the crust twisted and pushed it, forming the features visible.

Flakes of clay form almost as much of the mold surface as copper. Numerous fragments of the mold imbedded in the ingot are charred from the heat of the molten copper but interestingly, these seem to be spread along the perimeter of the ingot's surface. The flakes of clay around the center of the surface, however, exhibit little if any charring. What little charring is present is likely resultant from the heat of the molten copper igniting the organic materials used to temper the clay.

Sectioning the ingot revealed a unique pattern of gases trapped at its interior, with the majority of the gas pores located just beneath the rough surface. Their location here is not unexpected, since the clay mold only allows for the dissipation of soluble gases through the rough surface of the ingot, so long as the copper remains molten.

There is also substantial porosity near the mold surface, occurring in conjunction with a trapped piece of mold clay. The presence of this porosity, and the associated smaller pores, may have to do with the imbedded fragment of clay. It is possible that when pieces of clay flake off from the mold basin, it is accompanied by a corresponding release of gases from the mold. This situation would explain the additional porosity near the mold surface.

Interestingly, there are also small-sized pores that appear near the side of the ingot. The pores there are average in size and do not appear to be associated with any trapped clay particles. They are instead, likely the result of gases that merely became trapped within the more rapidly cooling copper around the edges of the ingot

Microscopic Observations

The second single-pour ingot cast in an open clay mold shares some features with the first, while still having an altogether unique appearance under magnification. Grain size is even smaller than what was observed in the first single-pour ingot cast in clay, ranging from 30 to 55 μm , with an average diameter of 40 μm . Porosity and inclusions of precipitates in grains are increased in this ingot to magnitudes that are unmatched among single-pour ingots. In fact, the presence of porosity, precipitates, and various inclusions is so extreme in this ingot that approximations of grain size near the rough and mold surfaces is nearly impossible. At this time, it can only be assumed that the grain size near the rough surface is smaller than that observed throughout the midsections of the ingot.

Porosity is pervasive in this ingot; the entire body is saturated with large and small pockets left by trapped gases. The areas bordering the rough and mold surfaces seem to exhibit the greatest amount of porosity, here, the porosity is not only more dense, but the largest pores occur there as well. As can be seen in Figure 14, both the upper and lower portions of the ingot contain large and round pores, as well as those that do not appear to have any discernable shape.

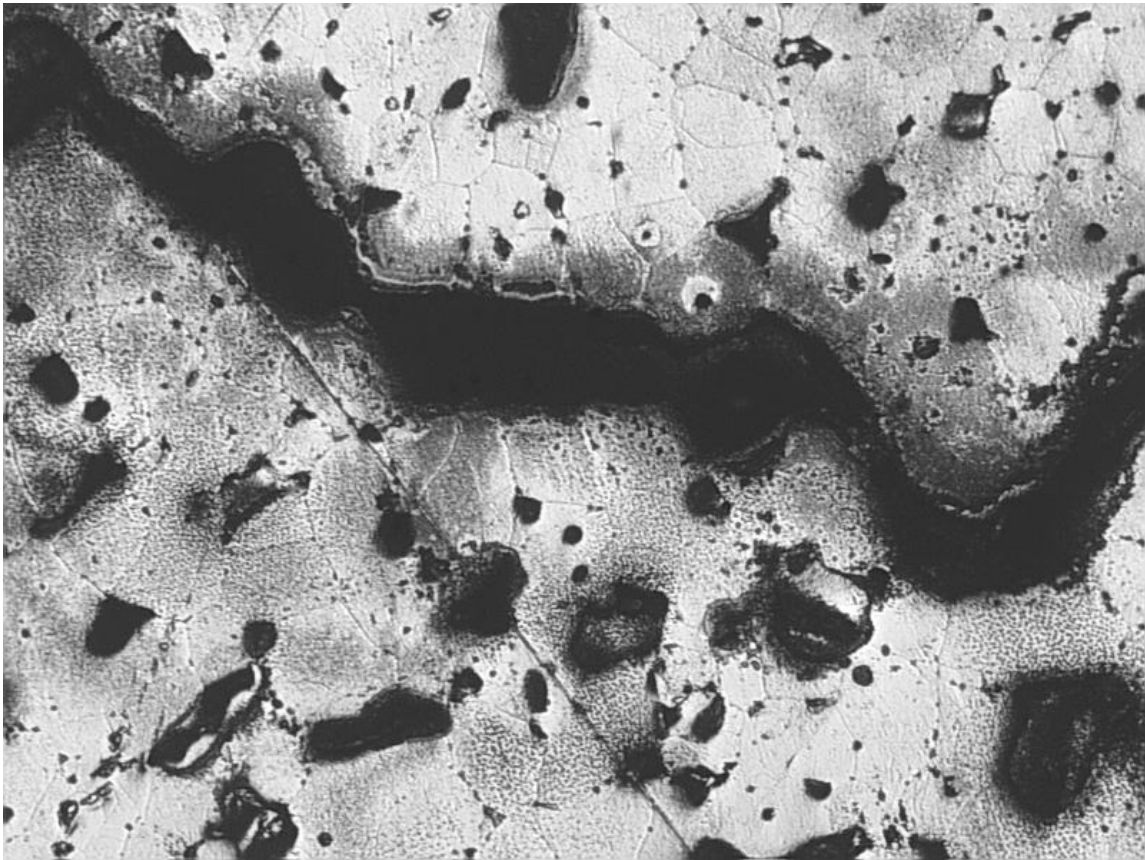


FIGURE 15. Porosity within the second clay mold, single-pour ingot at 200x magnification. Porosity in this ingot seems to have no uniform pattern. Also of note is the large crack dominating the center of the photo (photo by author, 2007).

Porosity in the mid-body of the ingot is somewhat different than what is observed in the upper and lower portions. While the number of pores is high, their size, for the most part, is smaller than that of the average copper grain. There are, however, exceptions to this and their presence is readily evident. Additionally, there is at least one example of a large crack in the copper matrix. Figure 15 shows clearly an elongated crack that is quite unusual and was not noted in any other ingot. This crack was likely formed as the ingot cooled and contracted. The edges of the gap left by this feature align with one another indicating that they were, in fact, joined at one time. In fact, there are many visible grain boundaries that are slightly larger, giving the appearance that the ingot cooled more rapidly and, therefore, the bond that fuses the copper grains together suffered damage.

In addition to the high frequency of pores within the copper matrix of this sample, there is an elevated number of precipitates. This is to be expected, given the relatively large amount of precipitates in the other single-pour clay cast ingot. Indeed, it is somewhat reassuring to find such similarity with respect to the amount and appearance of the precipitates in both ingots cast using the same procedures. Figure 16 clearly shows an increased frequency of precipitates along grain boundaries. It should be noted that many of the precipitates in this sample are significantly larger than those found in the other single-pour ingot cast in clay molds. Nonetheless, the similarities between the two single-pour ingots cast in clay molds are striking and significant and their resemblance to those examined from the Uluburun shipwreck is intriguing.

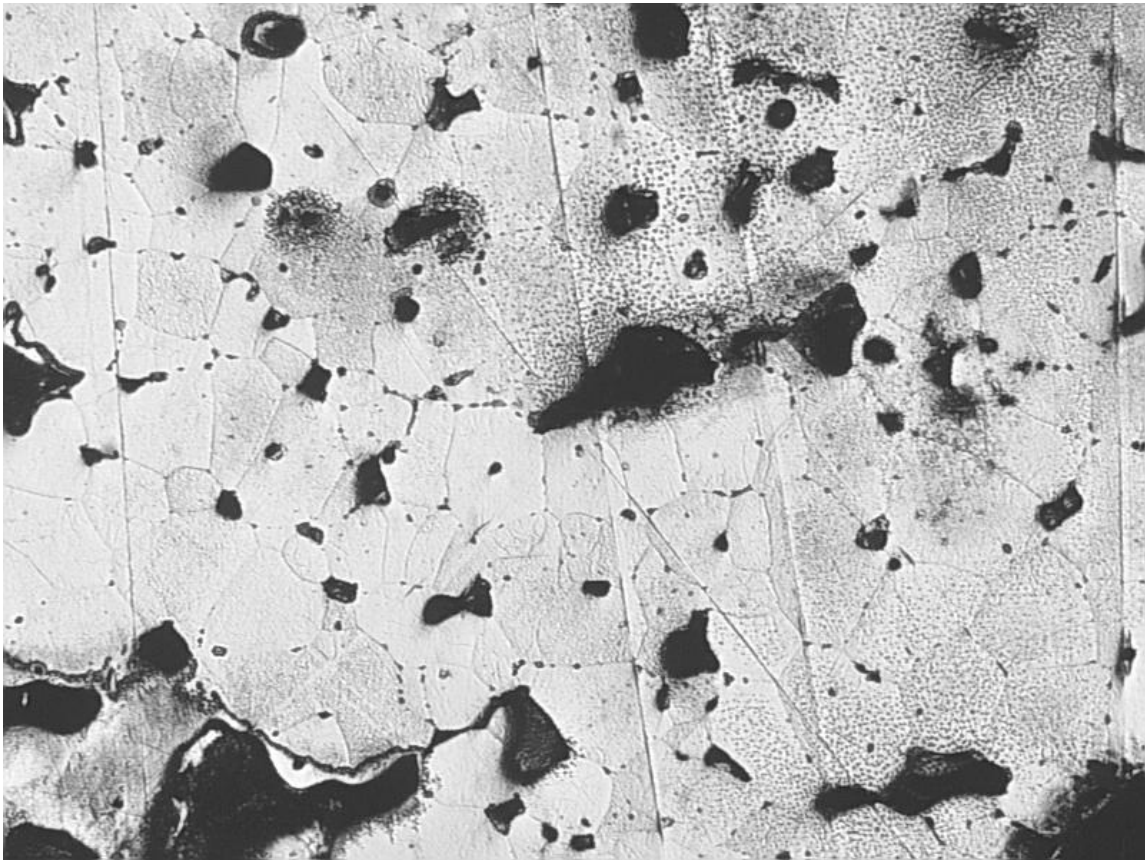


FIGURE 16. Second clay mold, single-pour ingot at 200x magnification. A high number of precipitates along grain boundaries are evident (photo by author, 2007).

Double Pour Ingots

Limestone Mold, Double Pour: Hot

Macroscopic Observations

This ingot was cast by pouring half the contents of a crucible into a limestone mold and allowing the molten metal to cease all reactive activity, and begin to transform from the liquid state to the solid state. Once this occurred, as indicated by the cessation of all bubbling of the molten copper and a thin film of metal copper forming on the surface, the remaining molten copper in the crucible was poured into the mold. The resulting limestone-cast, hot double-poured ingot weighs 4.5 kg, has a maximum diameter of 19.7 cm, a minimum diameter of 16.4 cm, a maximum thickness of 3.3 cm, and a minimum thickness of 2.4 cm.

The initial pour of copper resulted in significant bubbling and agitation, which likely resulted from the decalcining of the limestone mold and the effervescing of resulting carbon dioxide gas through the molten metal. The second pour of copper, made after a solid crust had formed on the surface of the first pour of copper (less than one minute between pours), again exhibited substantial bubbling, which was surprising since the portion of the ingot in contact with the mold (the first pour) should have cooled sufficiently so that the limestone no longer evolved gas due to its breakdown. Nevertheless, the second pour of copper bubbled much the same as the first pour. Upon cooling, a swell of metallic copper rose near one of the edges and formed a miniature volcano-like cone from which spewed molten copper. In one instance, the molten metal

flowed across the ingot forming what previously on the Uluburun ingots would have been termed a long “blister.” However, since the process by which this feature is formed is now known, those labeled as blisters on archaeological ingots, may more likely be the solidified copper that had oozed from a similar large bubble as the ingot cooled.

The rough surface of the hot-double-poured, limestone-cast ingot is covered in a thin, shell-like layer of copper. This thin crust of copper may be interpreted as a large blister, covering nearly the entire surface of the ingot. Upon visual inspection of the rough surface, the only area that does not appear to be covered by the large blister is a circular region that emanates from the volcano-like cone (Figures 17, 18). This portion of the rough surface appears to have a more solid foundation, rather than abundant gas cavities that lie beneath the blistery crust of the outer areas. The surface of the non-blistered region is noticeably smoother than the rest of the rough surface.



FIGURE 17. The formation of a volcano-like gas blister on the edge of an ingot cast in a limestone mold (photo by author, 2006).



FIGURE 18. The solidified gas blister on the surface of the ingot after cooling completely (photo by author, 2006).

On one side of the rough surface, the copper overflowed the mold during casting and there remains evidence of this event. Copper on the edge where it overflowed the mold protrudes out slightly and overhangs the side, like the upper part of a muffin or cupcake overhanging its paper wrapper. This feature should be evident on all ingots that overflowed their molds, since a thin layer of molten copper coming into contact with a cold mold would result in almost instantaneous solidification.

The mold surface of the hot double-poured, limestone-cast ingot differs significantly from the single-pour, limestone-cast ingot discussed earlier. While the single pour ingot has numerous gas pores on its mold surface, the hot, double-poured ingot displays significantly reduced porosity. What little porosity that is evident is spread equally throughout the mold surface, but the pores are substantially smaller in diameter and depth than those in the single-pour ingot cast in limestone. The reason for the lack of gas porosity in this ingot is likely due to the first pour of copper being only half an ingot's worth and, accordingly, the product was half as thin as a typical ingot. Since this resulted in a thin ingot, the soluble gases in the copper and the carbon dioxide created from the breakdown of the limestone mold had a shorter distance to travel and could, therefore, escape into the atmosphere more readily. Accordingly, if the gases are escaping more easily, then less will be trapped beneath the ingot's crust, resulting in reduced porosity.

There is, however, one distinctly large gas pore on the mold surface near the outer edge of the ingot. This pore appears to penetrate the first pour of the ingot, thereby offering a glimpse at the undersurface of the second pour. This interpretation, however,

is unlikely, and what appears to represent the underside of the second pour probably corresponds to the thin crust of copper that originally formed on the rough surface of the first pour. The large size of the gas cavity is unusual and unexpected, given how quickly the outer edges of an ingot cool, making the evolution of carbon dioxide in these regions less pronounced than that near the center of the ingot. Nevertheless, the line delineating the boundary between the first and second pours near the large pore, indicates the first pour was much thinner there than at any other location in the ingot. The cause of this imbalance is undoubtedly the result of the mold not being level during casting. Since the thinnest portion of the ingot would thus cool the fastest, it is reasonable to assume it would trap the most effervescing gases and perchance contain large cavities of gas and a high frequency of pores on the mold surface.

The mold surface of this ingot is distinctly marked by the roughness of the limestone mold in which it was cast. The abnormally rough mold surface is not a function of the copper's reaction with the limestone mold, but the result of a hastily made, uneven mold.

Given that this ingot was made from two pours of copper in rapid succession, a scrutiny of its side is of interest since it bears the features that mark the boundaries between the first and second pours of copper. Evident immediately, is a well-defined line that clearly indicates the two halves of the ingot associated with the successive pours. The line is extremely fine and sharp, and does not necessitate any speculation as to what it represents. The physical appearance of the boundary area indicates that despite the relatively short time lapse between the pours (less than a minute), the first pour

contracted as it cooled, and the molten copper from the second pour overlapped it slightly, and thus enveloped the upper portions of the first pour.

Microscopic Observations

Grain size and shape in this ingot is much the same as was observed in its single-pour counterpart. Grains are equiaxed and range in size from 45 μm to 130 μm , with an average diameter of 70 μm . As expected, grain size along the outer edges and top are slightly smaller, averaging approximately 35 μm , and rapidly increase in size toward the center (Figure 19).

Porosity is slightly increased in the ingot, especially in its lower portion. This is likely a result of the second pour of copper, which reheated the first pour, allowing more gases to come out of solution only to become trapped beneath the added layer of copper. While the single-pour ingot cast in limestone was relatively devoid of gas pores, this ingot exhibits slightly increased porosity. Even so, this ingot appears to be far more dense than any sample analyzed from the Uluburun shipwreck.

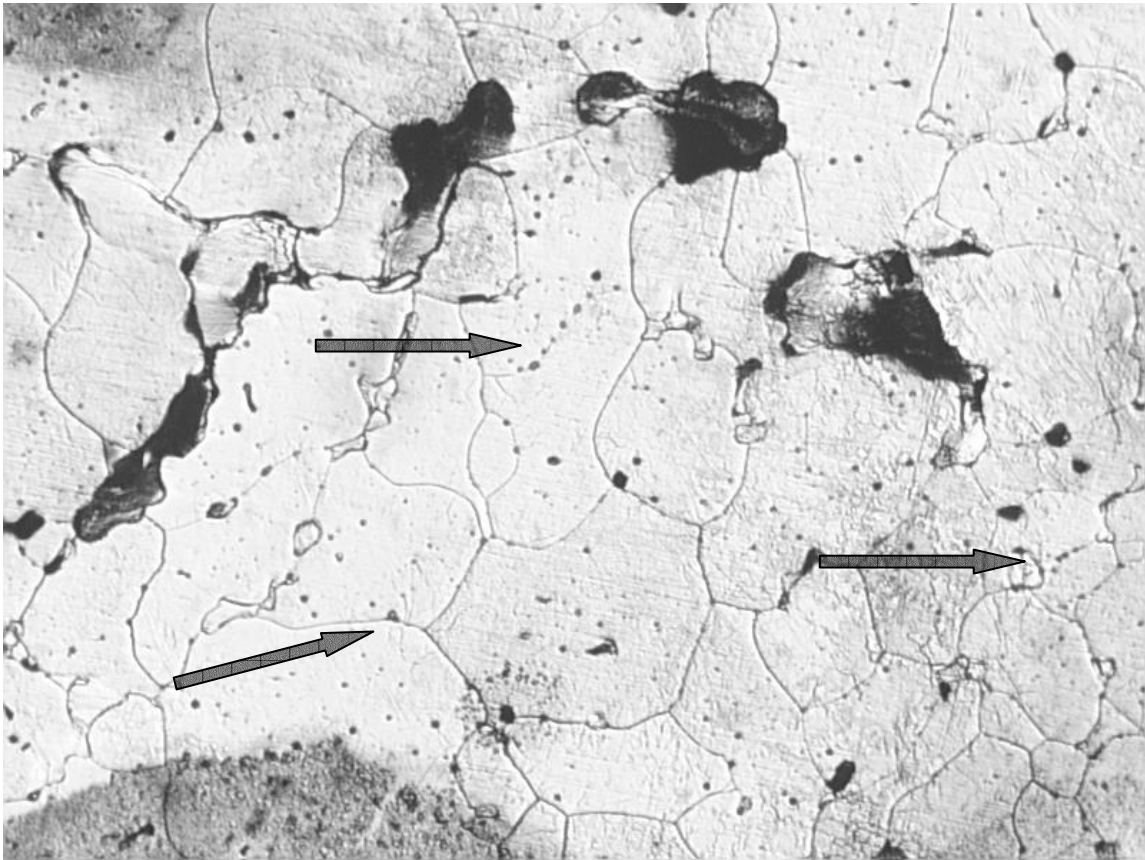


FIGURE 19. Grain details of the limestone mold, hot double-pour ingot at 200x magnification.

Intra-granular precipitates within equiaxed grains (photo by author, 2007).

While porosity was not significantly increased, the size and quantity of precipitates in this ingot was. The ingot still exhibits the small precipitates within the boundaries of the individual grains. One area of interest, highlighted in figure 19, may correspond to the boundary between the first and second pours of copper, and represent the oxide layer separating the two pours of copper. The second copper pour was not made until a thin crust formed on top of the first pour of copper. It is likely that the crust was formed mostly of oxides and other precipitates that did not mix with the pure copper grains. Therefore, the thin line that appears to form a partial barrier is likely formed by the remains of the crust that formed atop the first pour of copper. If this is correct, it would appear then that although the two copper pours fused together completely at the time of casting, microscopic examination indicates otherwise (Figure 20).

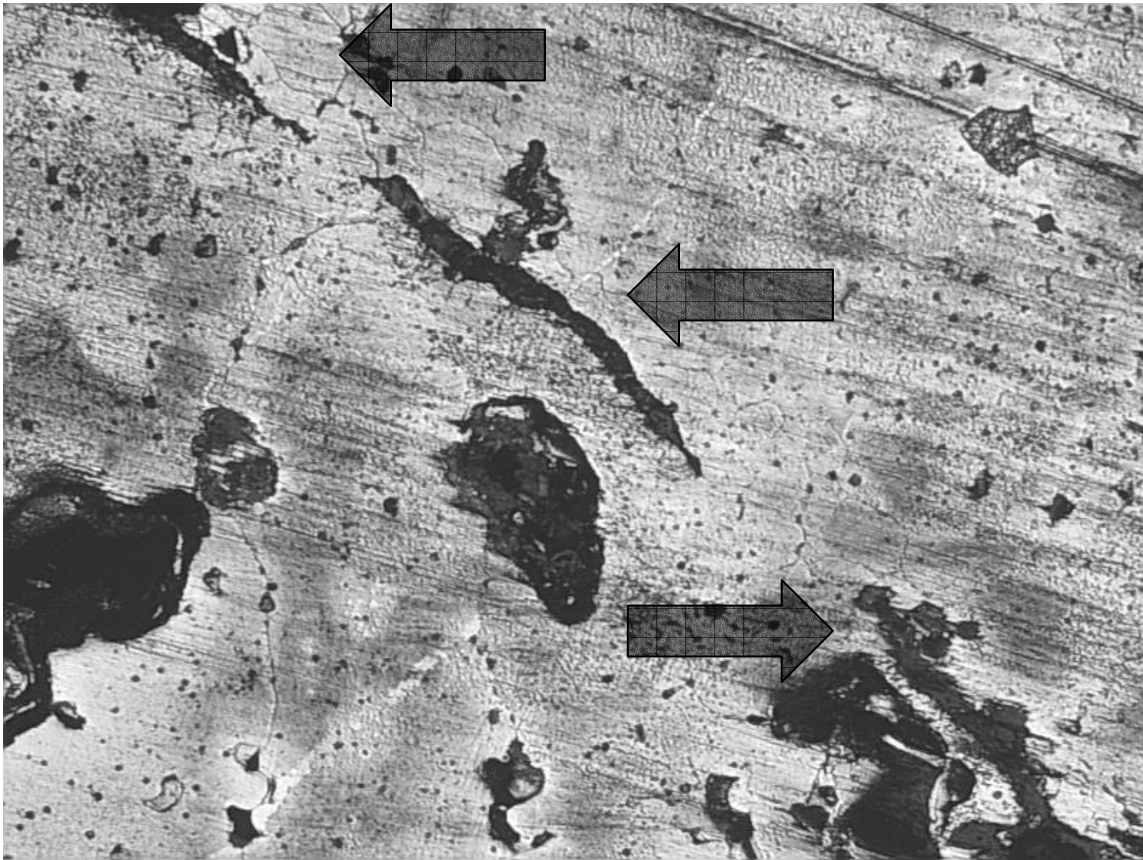


FIGURE 20. Limestone mold, hot double-pour ingot at 200x magnification. The series of elongated voids running diagonally from top left to bottom right may represent the boundary between the two pours of copper (photo by author, 2007).

Limestone Mold, Double Pour: Cold

Macroscopic Observations

This ingot, resulting from two pours of copper—the second made after the first had cooled to the touch—has a maximum diameter of 20.1 cm, a minimum diameter of 17.3 cm, a maximum thickness of 3.5 cm, a minimum thickness of 2.8 cm, and a weight of 5.44 kg. The ingot displays a unique rough surface compared to the other ingots cast in this study. While the rough surfaces of ancient ingots, and those cast thus far for the purposes of this study, have been coarse and bubbly, the rough surface of this ingot has a relatively smooth consistency. This is not to say there are no irregularities, but rather that the appearance of the ingot's top surface has almost a milky quality. Near the center of the ingot there is a substantial cold shot, where the last bit of molten copper was poured from the crucible onto an already cooling and non-reactant body of metal. The cold shot is in the form of a smooth glob of copper that appears to have been added as an afterthought. This feature demonstrates how fast molten copper cools when poured onto a metal surface at ambient temperature.

It appears that the second pour of copper, comprising the upper half of the ingot, and therefore including the rough surface, cooled so rapidly that it did not fully cover the first pour of copper and portions of the first pour remained visible. There are even clear indications of where the molten second pour of copper flowed around some of the more prominent portions of the first pour—for example, a tall volcano-like blister—but failed to cover them completely. From this, one can see the direction of copper flow and can,

therefore, infer the general location from which the copper was poured into the mold. Further support for such inferences comes from the shape of the previously discussed cold shot. The edges of the cold shot plainly illustrate the direction in which the copper flowed across the surface as it cooled.

A curious feature on this ingot can be seen on both the rough and mold surfaces. Near the edge of these surfaces, there are distinct cracks in the second pour of copper. The cracks are likely the result of the second copper pour cooling and contracting around the already formed first pour. Obviously, this feature could not have occurred in the initial stages of the cooling, and its formation must have taken place after the second pour of copper pour had already solidified into shape. As the second pour cooled, it would have contracted as it went transitioning from liquid to solid metal. As the copper cooled it would have continued to contract further, and since portions of the second pour encompassed the first, the copper cracked as it contracted and stretched tight around the already solidified first pour.

The most conspicuous feature on the ingot's mold surface is the copper from the second pour, which seeped around the already cooled first pour of copper, as it was poured. Aside from its metallic sheen, the copper that oozed into the mold gaps formed by the contraction of the first copper pour bears some resemblance to melted candle wax. Long fingers of copper (Figure 21) from the second pour stretch across approximately $1/5^{\text{th}}$ of the mold surface. Due to their thinness and relative separation from the bulk of the second pour of copper, many portions of this underflow, exhibit the cracks associated with cooling discussed above.



Figure 21. Limestone mold, cold double-pour ingot. Note the copper tendrils on the mold surface of the ingot (on the top of photo) and the distinct separation between the first and second pours of copper (photo by author, 2006).

Upon inspection, there do not appear to be any gas pores present on the mold surface of this ingot. This is contrary to the appearance of the single-pour ingot cast in limestone, but parallels the results from the hot double-poured limestone-cast ingot and the two preliminary ingots deemed too thin for comparative results. As previously hypothesized, it is likely that the lack of porosity in the first pour—which corresponds to the formation of the mold surface—is due to the thinness of the initial casting. A smaller volume of copper allows inherent gases and those created from the breakdown of limestone to escape more easily, thereby reducing pores formed by trapped gasses on the mold surface.

Initial sectioning of this ingot for microscopic analysis revealed some unique and unexpected features. As noted above, the copper from the second pour seeped around the edges of the first pour, solidified, and became a conspicuous feature on the mold surface. Cutting into the ingot revealed just how drastic and encompassing that overflow was, as it became clear how the first pour of copper was completely engulfed by the second pour. Aside from the porosity that formed around the boundaries between the first and second copper pours, the ingot appears to be relatively dense, exhibiting only a few pockets of trapped gas.

Microscopic Observations

Despite the relatively dense appearance of this ingot at the macroscopic level, microscopic examination reveals a structure that is quite different than what was

observed in the other two ingots cast in a limestone mold. Although the lower and upper ingot halves seemed to have bonded together reasonably well, cutting sections for sampling revealed just how weak these bonds were, as the two sections easily separated and had to be examined individually under magnification.

Since the first pour of copper was, in reality, a separate ingot prior to the addition of the second pour, it would stand to reason that it would share similar features to the single-pour ingot cast in limestone. This reasoning proved to be correct, as this half of the ingot indeed compares fairly well to its single-pour cousin. As with the single-pour ingot, this ingot was also found to be dense, with few pores. Additionally, there are substantially fewer precipitates in this ingot compared to those examined from the Uluburun shipwreck. As with the single-pour, limestone-cast ingot, what few precipitates observed are primarily within the borders of the individual copper grains, rather than concentrated at the grain boundaries, as is observed with the Uluburun ingot examples. The copper grains in this sample are equiaxed, but are significantly smaller than those from the single-pour ingot, ranging in size from 40 μm to 80 μm , and averaging about 50 μm . Near the top surface of this ingot half, the grains size is reduced even more substantially (Figure 22).

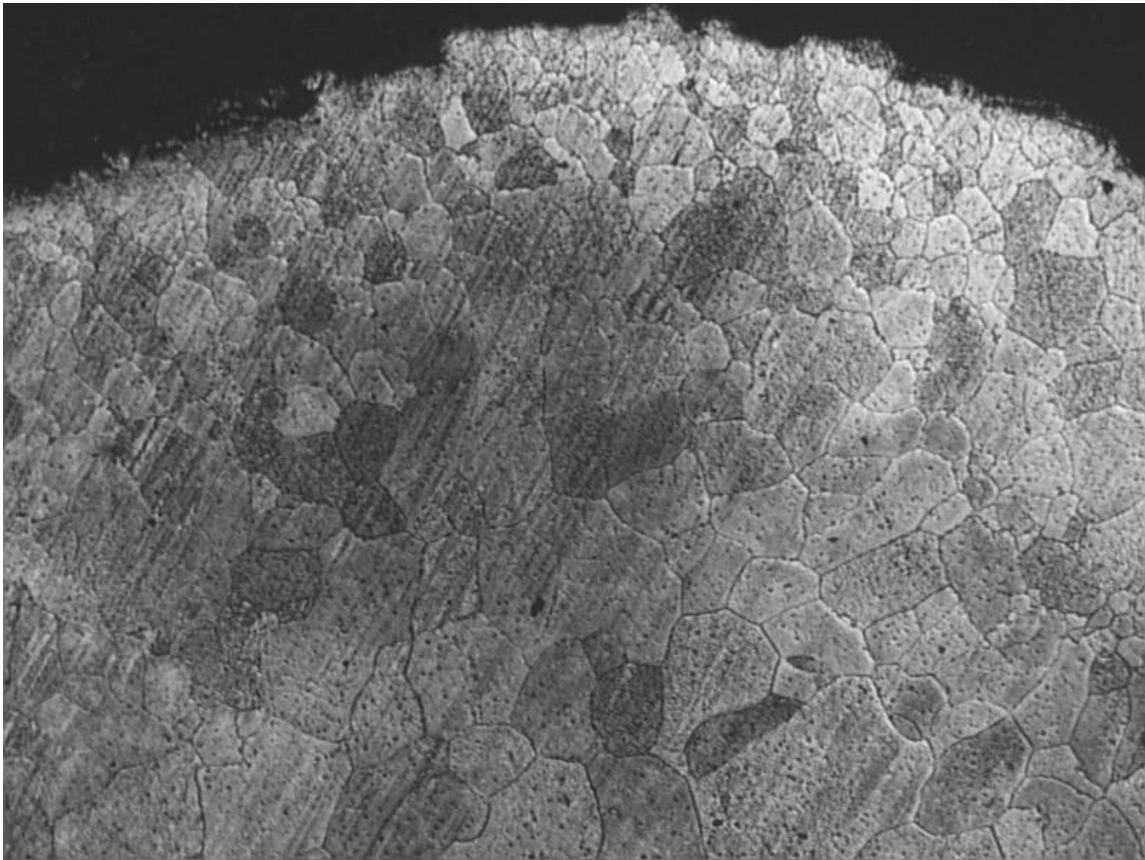


FIGURE 22. Grain details of the limestone mold, cold double-pour ingot at 200x magnification.

Notice how drastically grain size reduces approaching the upper portion of the ingot. Note the drastic reduction in grain size near the top surface of the ingot due to more rapid cooling (photo by author, 2007).

The reduced grain size of the first pour, when compared to the single-pour ingot cast in limestone, is likely due to its decreased thickness. Since this half of the ingot represents the first of two pours intended to create a full-sized copper ingot, it follows that it is only half as thick as an ingot consisting of a single pour. Since the volume of metal in this half of the ingot is only half that of a full-sized ingot, it will dissipate heat quicker than an ingot twice its thickness. Consequently, the grain size would be smaller, since it would have cooled much quicker than the single-pour ingot cast in limestone.

The upper half, or second pour, of this ingot is quite different from the lower one. The most significant disparity between the two halves comprising this ingot, is that it is nearly impossible to move the microscope over the surface of the sample without encountering numerous large pores. Undeniably, the second pour of copper is far more porous than the first, and the reasoning behind this is not so difficult to perceive. As evidenced by the previous two ingots cast in limestone, this particular mold material is semi-porous and allows for limited escape of soluble gases in all directions, not just through the ingot's exposed upper surface. The second pour for this ingot, however, was made on top of a solidified, non-porous surface formed by the first pour of copper. Given that hardened and cooled copper is not a material that will allow for the diffusion of soluble gases, those gases that would have normally escaped through the bottom of an ingot and out through the mold, remained trapped in the copper matrix. The process of trapping gases was exacerbated by the fact that the second pour of copper was also thin and therefore cooled rapidly, decreasing the chances of the effervescing gases escaping.

Confirmation of this can be seen in the photos of the ingot taken through the microscope showing increased porosity.

The grains of the second pour of copper are not equiaxed, varying in size and shape throughout the body of this ingot. With respect to size, grains vary from 20 μm to 90 μm and seem to average about 50 μm in diameter. The smallest grains are found near the rough surface and along the boundary with the first pour, while the larger grains are localized in the center of this ingot half.

In addition to significantly increased porosity, the second pour of copper contains a greater amount of inclusions and precipitates as well. The highest concentration of precipitates seems to be located near the border between the first and second pours. There, the grains are most easily recognized by the precipitates concentrated along their borders. In fact, in Figure 23, it appears that most copper grains are delineated by a border of precipitates. The small grain size and high incidence of precipitates along the lower edge of the second pour of copper are likely due to the molten copper being poured on top of an existing layer of already cooled copper. Since the specific heat of copper is lower than that of limestone, the first pour of copper would have absorbed the heat from the second pour more readily than the limestone mold could have absorbed the heat from the first pour of copper. Thus, the second pour of copper would have cooled more quickly than the first, thereby rendering a smaller more tightly packed matrix of copper grains.

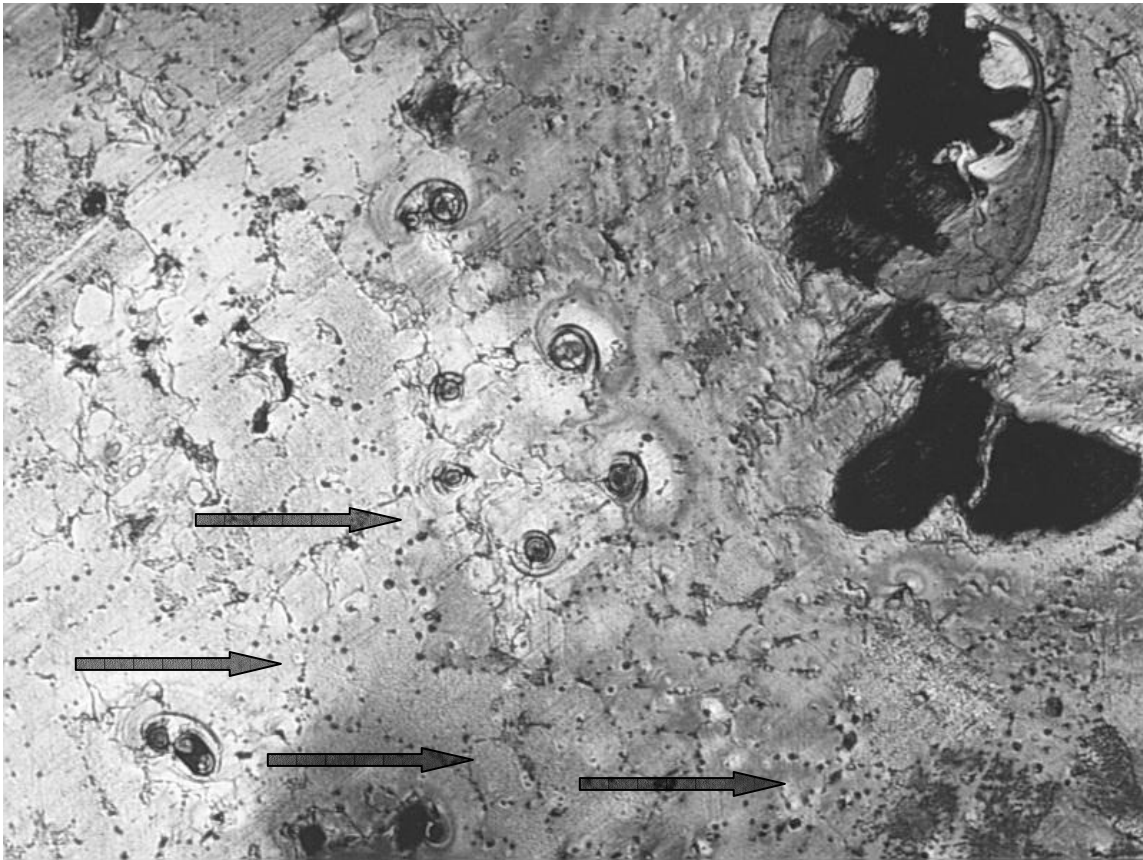


FIGURE 23. Limestone mold, cold double-pour ingot at 200x magnification. The grain boundaries are difficult to discern, but are delineated by precipitates (photo by author, 2007).

Sand Mold, Double Pour: Hot

Macroscopic Observations

Two multiple pour ingots were cast in a sand mold to test different multiple pour casting techniques. The first ingot, the double-pour hot ingot was produced with two pours of copper done in close succession. The second pour of copper was made as soon as a thin crust had formed atop the cooling, still molten, ingot. The second ingot, the double-pour cold ingot, was produced with two pours of copper separated by several hours time. In this instance, the second pour of copper was made only after the first had cooled to the ambient temperature of the environment.

Unlike the hot double-pour ingot cast in limestone, the boundaries between the first and second pours of this ingot are much less distinguishable. Along the sides of the ingot, the boundary is defined by small flashes or fins of copper and slight depressions or grooves. Additionally, there appears to be a color change corresponding to the two separate pours. The bottom half of the ingot (first pour) is gray in color, while the upper half (second pour) has a distinctly brown hue. The color difference is difficult to explain and could have resulted from materials used in the mold or from the speed at which the two pours cooled. The grayness of the first pour may have resulted from the adherence of gray bentonite clay to the ingot, while the brown coloration could have been caused by sand adhering to the ingot. Or, as postulated above, the grayness of the first pour could be the product of relatively slow cooling, while the brown hue of the second pour may result from relatively quicker cooling. At present, there is no obvious explanation

for why more bentonite clay seems to have stuck to the first pour, while the second pour appears to have attracted more sand. Aside from the color differences and localized slight depressions and fins, there is not a clear and definable seam between the first and second pours of copper, as have been observed on the limestone-cast ingot of the same type.

The rough surface of this ingot appears to be made up of a thin crust of copper, which is likely the result of rising gases that could not escape before the surface hardened. This phenomenon—trapped gases beneath the “crust” surface—was observed in every copper ingot. Since it is exposed to the air upon casting, the uppermost surface of an ingot cools the fastest. This quick cooling of the rough surface forms a thin crust of solid metallic copper over the molten copper, which, in turn, effectively traps any gases seeking escape. With the formation of the crust, one can actually observe gas bubbles collecting beneath its surface. They appear much like small bubbles of air floating on water beneath a thin layer of ice. The trapped gasses appear as dark, moving bubbles beneath the thin crust of the rough surface, which contrast sharply with the glowing orange of the still molten copper on which they float. These gas bubbles, which grow and sometimes push the crust upwards causing it to rupture, are the probable cause for the thin crust observed on the rough surface above the more solid corpus of an ingot.

Near the center of the rough surface are several prominent blisters that flank a larger swell. These blisters are not products of engorged gas bubbles, but molten copper that erupted from the swell in the center of the ingot. As an ingot's surface cools and trapped gases build up below it, the crust of the rough surface swells and cracks,

allowing molten copper to ooze out onto and across the newly hardened surface. These blisters are themselves porous and exhibit evidence of trapped gases.

As mentioned above, the rough surface is covered with a thin crust of copper that conceals many cavities and the more solid body of the ingot. The crust of the rough surface is relatively smooth around its perimeter when compared to the swells and blisters near the center of the ingot. This is probably caused by the quicker cooling of copper in the regions of the surface, resulting in the perimeter surface being made of smaller copper crystals than those that make up the slower cooling center of the ingot.

Present on the rough surface are several coal and sand inclusions resulting from the casting process. The rapid deterioration of the furnace lid used to melt the copper resulted in fragments of the refractory material, consisting of cement and sand, falling into the crucible, which found their way as intrusive features in the final casts

The mold surface is typical of the sand-cast ingots. The entirety of the surface is covered with bits of bentonite clay particles transferred from the mold. The clay particles are so pervasive that it makes locating and identifying features of the mold surface difficult. Also present on the mold surface is one semi-prominent swell and two smaller swells. The most visible of the three swells is likely the result of loosely packed sand on the bottom of the mold being displaced by molten copper at the time of casting.

Sectioning the ingot for examination of its interior reveals that it is quite dense, much like the single-pour ingot cast in a sand mold. Examples of visible pores are evident very near the mold surface, where a concentration of very small pores have formed along the lower boundary. Additionally, there are three notable pores roughly

midway through the interior of the ingot. Their location could correspond to the original rough surface, upon which the second layer of copper was poured. Lastly, the only other examples of trapped gasses in this ingot are found just below the rough surface, where they were likely prevented from escaping by the formation of the initial thin crust as the ingot cooled.

As with the exterior of this ingot, there does not appear to be much evidence for the multi-pouring of the ingot in its interior. The only indicators that are visible to the naked eye may be the three pores near the midline of the ingot, representing evidence of gases trapped beneath the initial crust of the cooling copper, prior the addition of the second pour.

Microscopic Observations

The microscopic structure of this ingot does not reveal any more about its two-pour formation than did the visual examination. In fact, the only observable difference between the single pour ingot and this one, is that the latter contains significantly more precipitates. Copper grains are equiaxed and their size remains roughly the same as in the single-pour ingot cast in sand, and ranges from 60 μm to 100 μm . A striking difference between this ingot and its single-pour cousin, is that the grains of this ingot do not appear to contain the directionality of the single-pour ingot, which is odd, since they should have both cooled at roughly the same rate. Tentatively, this anomaly could be explained by the introduction of the second pour of copper. Perhaps, introducing another

pour of still molten copper to the cooling ingot could have potentially reheated it enough and, in effect, increased the cooling time enough to keep the copper grains from freezing in a directional manner.

Sand Mold, Double Pour: Cold

Macroscopic Observations

The double-poured ingot cast in sand, where the second pour was made over a cold ingot, exhibits at least one feature on the rough surface that appears to be unique among the ingots cast during these experiments. The rough surface of this ingot displays an unusually high frequency of small gas pores. On both the Uluburun ingots and those created in these experiments, gas porosity—especially those of this nature and of this quantity—are rare on the rough surface of an ingot. Indeed, blisters and blows do form on the rough surface of ingots, but numerous tiny pores have yet to be documented. Tiny gas pores are the type of feature one expects to find on the mold surface of an ingot, but not on the rough surface. The pores are larger near the edges of the rough surface, decreasing in size but remaining equally plentiful around the center of the ingot. The only portion of the rough surface devoid of gas pores is a small area of smooth copper near one of the edges. It is probable this is the area where the copper was poured and was therefore last to cool on the ingot.

Other than the abundance of gas pores on the rough surface, additional features observed include the ubiquitous blisters and coal inclusions. There does not seem to be

any pattern to the blistering on the rough surface, as the blisters appear to occur at a random frequency, as do the pieces of coal scattered about the ingot's surface.

The mold surface of this ingot contains a prominent and uncommon feature. Approximately midway through, and nearly bisecting the surface in two portions of unequal heights, runs a sharp ridge. Upon closer inspection, this feature does not appear to be a ridge at all, but evidence of copper that overlapped and seeped beneath the original pour of copper. Since this is a two-pour ingot, a feature such as this would not normally seem unusual. What is unusual is that the overlapping does not appear to be connected with the second pour of copper, but rather is a result of the first. It is also possible that this feature resulted from a defect in the mold and is not an overlap of the second pour. What appears to be a thin run of copper over a cooled surface may in fact be nothing more than a slight seeping of copper around loose sand in the bottom of the mold. The exact cause and description of this feature cannot be determined with certainty without slicing the ingot in two halves. Unfortunately, the machinery available for sectioning the ingots did not allow a cut to be made that far into the center of the ingot.

Other than the unusual ridge on the mold surface, the only other feature readily apparent is a small swell near one edge. Again, much like the other two ingots cast in sand, the mold surface of this ingot is covered with tiny bentonite clay particles. Moreover, there do not appear to be any gas pores on the mold surface.

As expected, the sides of this ingot bear traits that reveal the method used in its casting. Along the entire side of the ingot, copper from the second pour ran over the

edges of the first pour and seeped down and around it. In some instances, copper from the second pour flowed all the way to the bottom of the mold surface. However, for most of the ingot, copper merely ran over the edges and part-way down the sides. As with the other two-pour ingot, where the second pour was added to a cold lower ingot, there appears to be significant bonding between the two successive pours of copper. This is evidenced by the solid nature of the ingot and how the two halves do not move or jiggle.

Sectioning this ingot indicates that although the ingot halves feel well integrated, there is a definite line dividing the two pours. The interior of the bottom half of the ingot, as expected, resembles the other two ingots cast in a sand mold. The first pour is nearly devoid of pores, except for very near the mold surface, where small bands of pores have accumulated.

The dividing line between the two ingot halves is clearly evident and is emphasized by the presence of several large gas pores. The upper half of the ingot seems to contain significantly more pockets of trapped gas, all with distinct upward flowing, column-like appearances. Their columnar look is reminiscent of the grain structure of the single-pour ingot cast in sand, and likely resulted from the more rapid cooling of this thin section of copper.

Although joined together, the dissimilar look of the two pours of copper make it readily apparent that this ingot was cast in two separate pouring operations, with a substantial amount of time separating the two copper pours. While the two halves do comprise a single unit, it appears as if this specimen consists of two distinct and different ingots.

Microscopic Observations

Although the two halves of the ingot appear quite different to the naked eye, at the microscopic level they are not all that dissimilar. Both halves of the copper ingot exhibit equiaxed copper grains that are significantly smaller than those observed in the other two ingots cast in sand. The grains vary in size from 30 to 60 μm with an average size of about 45 μm in diameter. As expected, grain size is greatly reduced near the tops and bottoms of the two halves, due to the increased cooling rate of the copper at these locations. From the sections examined under the microscope, it appears that the lower half (first pour of copper) is actually more porous at the microscopic level than the upper half. The cause of this is likely related to the increased macroscopic porosity in the upper half of the ingot. If the copper had stayed molten long enough for the trapped gases to gather into larger bubbles, then perhaps there would have been less microscopic porosity in the upper half of the ingot compared to its lower half.

Both the upper and lower portions of the ingot contain large quantities of precipitates. In the lower half, or first pour, the majority of precipitates are found in large pockets resembling gas pores. These pockets of material extend along the boundaries of several grains. In addition to the larger precipitates, the lower half of the ingot contains some smaller precipitates that seem to congregate at the junction of grain boundaries.

The second pour of copper (upper half of the ingot) seems to contain more of the smaller precipitates, which in places almost seem to define the grain boundaries. Despite

the increased number of smaller precipitates, the second pour of copper also contains some of the larger precipitates, although not as much as in the lower half.

Clay Mold, Double Pour: Hot

Macroscopic Observations

The rough surface of this ingot has a central feature that is unique among all of the ingots produced in these casting experiments. Near the center of the rough surface is a fissure or valley-like feature caused by swelling, due to rising gases within the molten cooling molten metal. The feature undoubtedly resulted from trapped gases rising in the ingot after a thin crust had formed on the rough surface. As pressure from the trapped gases built against the crust, the rough surface rose to accommodate the gas expansion until the crust finally burst open, allowing gases and molten metal to gush forth and flow over the cooling surface of the ingot. The running molten copper then piled up against the side of the clay mold, creating a large and distinct fin. The swell in the center bordering the fissure contains numerous blows resulting from the gas pressure once held underneath the surface.

Aside from the swell and fissure in the center of the ingot, the rough surface exhibits the common small blisters and blows that are typical of ingots from antiquity and those created in these experiments. Portions of the rough surface not containing blisters and blows, cooled in a manner revealing the formation of a thin crust on top of still moving molten copper. In these places, the rough surface still bears the appearance

of a thin crust wrinkled and crumpled by the forces of shifting gases and flowing metal. Features such as this serve as a stark reminder that the observable attributes of ingots were created from the dynamic interactions of molten copper with the molds in which it was poured and the gases created from those interactions, as well as those liberated from the copper itself.

The rough surface of this ingot also contains a few small clay inclusions, resulting from the breaking down of the mold. There are not as many clay fragments on the ingot as there are on the first ingot cast in this clay mold. This is no doubt the result of the mold being more tolerant to thermal shock on its second use, having most of the areas susceptible to heat stresses destroyed during the first casting.

The mold surface contains more clay inclusions than the rough surface. This is to be expected, since that is the surface of the ingot in contact with the clay mold. The clay inclusions are clustered around the center of the mold surface, which is not surprising given that this was the hottest portion of the ingot during casting and cooling. The hottest portion of the ingot would naturally have imparted the greatest damage to the surface of a mold of any material. Many of the clay flakes imbedded in the mold surface bear the telltale signs of charring from the extreme heat of the molten copper.

Gas porosity is limited on the mold surface. What few pores are present are shallow and appear to result from small amounts of gas trapped between molten copper and the surface of the mold. There is no discernible pattern for these pores, as their presence is randomly distributed over the mold surface of the ingot.

Since the ingot was created by two pours of copper instead of one, the side of the ingots contains a faint line that reveals this aspect of its formation. The second pouring of copper was made as soon as a thin crust of metallic copper formed on the surface of the molten ingot. As a result of the short time interval between the two pours, the two halves are well integrated and there is no noticeable movement or jiggling between them. Nevertheless, a discernible line where the second pour joins the first is clearly visible along the side of the ingot.

Sectioning the ingot with a metal-cutting band saw revealed interesting features of its macroscopic internal structure. Given the numerous blows and blisters visible on the rough surface of the ingot, one would have argued that the body of the ingot contained gas pores as well. Examination of the sectioned ingot, did, indeed, reveal a distinct porosity in its inner structure. However, there are some observations that can be made with respect to the nature of the porousness of this ingot.

A quick glance reveals that the majority of the trapped gases are located near the mold surface of the ingot and tiny bubbles of trapped gases line the edges near the mold surface. These pockets of trapped gases grow in size toward the center of the ingot and become noticeably more infrequent near the rough surface. The upper portions of the ingot are not devoid of trapped gas pockets, but those that do appear are smaller in size and less abundant than those found near the middle of the ingot.

The placement of the trapped gases within the ingot are easily explainable by its production using two subsequent pours of copper. As the first pour of copper began to cool and gases contained in the molten metal tried to escape from the solution, they

naturally rose toward the surface and some of these gases escaped. However, since the second portion of copper was poured over the first as soon as a thin translucent crust formed on the surface of the molten ingot, those gases that had not yet effervesced became trapped beneath an even greater amount of copper. Given that the second pour of copper was made over an already hot metal surface, it is likely to have stayed molten longer, therefore, allowing a longer opportunity for the gases to escape than was afforded those effervescing from the first pour.

Microscopic Observations

Microscopic examination of this ingot reveals a structure that falls somewhere between those of the two single-pour ingots produced using a clay mold. Copper grains are equiaxed, varying in size from 50 μm to 70 μm in diameter. This ingot contains more gas porosity than normal, a feature that may be a function of the mold material, since the other ingots cast in clay seem to display above average porosity.

In addition to gas porosity, there is also a significant level of precipitates, large and small, contained within the copper matrix of the ingot. Some of the precipitates are located within the grain boundaries, but most seem to have gathered between the grain boundaries and at the intersections of several grains. Indeed, there may also be inclusions from the mold in which the ingot was cast, as shown by the variety of non-cuprous materials in Figure 24.

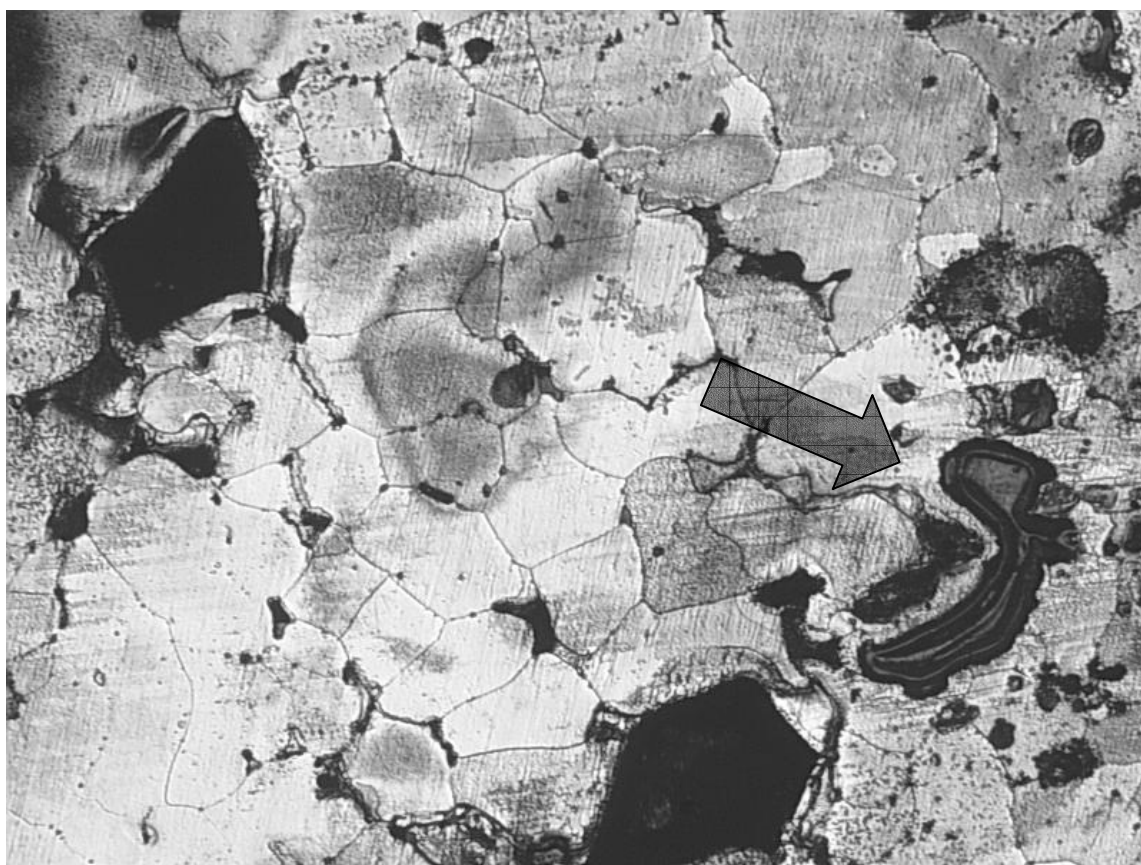


FIGURE 24. Grain structure of the clay mold, hot double-pour ingot at 200x magnification. The banana-shaped inclusion (lower right) may be a fragment of the clay mold (photo by author, 2007).

The composition of the ingot seems to change toward the mold surface. In addition to the smaller size of the grains at this location, the precipitates and the porosity seem to take on drastically different shapes than those observed throughout the remainder of the ingot. Both the precipitates and porosity near the mold surface seem to have much more globular, or round, appearances. In addition to the differing shapes of the inclusions at this location, they exhibited an increased abundance. The radical disparity between the inclusions in the body of the ingot and at the mold surface does not appear to be an exclusive feature of this ingot. Examining the microstructure of the second single-pour ingot cast in clay reveals similar formations near the mold surface. It would seem that the more rapid cooling and close proximity to the mold surface resulted in a microstructure unique to clay molds, which could prove to be a useful diagnostic tool when examining archaeological ingots in the future.

In order to summarize the products of the casting experiments, Tables 1 and 2 show the sizes and weights, as well as the grain structures, of all of the ingots cast in these experiments.

Table 1:
Ingot Sizes and Weights

	Diameter (cm)	Maximum Thickness (cm)	Minimum Thickness (cm)	Weight (kg)
Sand Mold; Single Pour	17.7	3	1.4	4.26
Clay Mold; Single Pour #1	18.1-16.8	3.8	2.9	3.72
Clay Mold; Single Pour #2	18.2-17.4	3.2	2.8	4.0
Limestone Mold; Single Pour	18.7-16.4	3.4	2.0	4.40
Limestone Mold, Double-Pour: Hot	19.7-16.4	3.3	2.4	4.50
Limestone Mold, Double-Pour: Cold	20.1-17.3	3.5	2.8	5.44
Sand Mold, Double-Pour: Hot	17.7	3.3	1.8	4.38
Sand Mold, Double-Pour: Cold	17.9	3.6	2.4	4.76
Clay Mold, Double-Pour: Hot	18.5-17.8	4.0	2.3	4.52

Table 2:
Ingot Grain Structures

	Grain Size (μm)	Grain Structure	Precipitates
Sand Mold; Single Pour	30-80	Not Equiaxed	Within grain boundaries
Clay Mold; Single Pour #1	30-80	Equiaxed	Within and along grain boundaries
Clay Mold; Single Pour #2	30-55	Equiaxed	Along grain boundaries
Limestone Mold; Single Pour	60-140	Not Equiaxed	Within grain boundaries
Limestone Mold, Double-Pour: Hot	45-130	Equiaxed	Within grain boundaries
Limestone Mold, Double-Pour: Cold	20-90	Equiaxed	Within grain boundaries
Sand Mold, Double-Pour: Hot	60-100	Equiaxed	Along grain boundaries
Sand Mold, Double-Pour: Cold	30-60	Equiaxed	Along grain boundaries
Clay Mold, Double-Pour: Hot	50-70	Equiaxed	Along grain boundaries

CHAPTER V

DISCUSSION AND CONCLUSIONS

Both the macroscopic and microscopic analysis of the experimental ingot samples reveal many characteristic aspects for each mold type, and help put the casting experiments into perspective. Conceivably, the most important aspect of the analysis stems from observations made of the ingots produced with a single pour of copper. The single-pour ingots reveal how each different mold material creates unique physical characteristics and microstructures for the ingots formed therein. The behavior of cooling copper in each of the three mold types used creates premises upon which speculation on ingot casting can be made. These premises, based on the fundamental manner in which copper reacts with each mold type, will hopefully lead to conclusions made about the mold materials used to cast copper ingots in the Late Bronze Age.

Examining the sectioned portions of all the ingots cast in these experiments reveals much as to how the properties of the different molds allow for the effervescence of soluble gases, whether they originate from the mold material or are found within the raw copper. One noteworthy observation is the lack of porosity in the ingots cast in a single pour, regardless of the mold material used. The ingot cast in sand is near perfect with respect to its density, with only slight porosity occurring first below the rough surface. The density of the limestone-cast ingot follows close behind its sand-cast counterpart, exhibiting only a few pockets of gas near the center of the rough surface, but remaining for the most part free of imperfections. The two clay-cast ingots exhibit

the most porosity resulting from trapped gases. The first clay-cast ingot, which was lens shaped in sectional view, displays a significant amount of small pores throughout its corpus, and one large gas pocket near the rough surface. The second clay-cast ingot is significantly denser than the first, with only tiny gas pores near the mold surface and a few larger ones near the rough surface. Much like the ingots cast in sand and limestone, the body of this ingot is largely devoid of porosity. However, the presence of trapped gases in the single-pour ingots cast in clay and the lack of pores in those cast in limestone and sand molds seem to implicate clay as a mold material less favorable for allowing the release of soluble gases as the molten copper cools and solidifies.

Beyond the apparent observation that single-pour ingots yield the least porous ingots, and that limestone and sand molds appear to allow gases to escape molten copper more readily than those of clay, there is difficulty in determining any patterns with respect to casting methods and/or mold materials. Judging strictly on the basis of single pour ingots, it would be easy to proclaim that ingots cast in sand are the least porous, since sand provides the least restricted passage for effervescing gases during the cooling of the ingots. Does this statement hold true for ingots cast in multiple pours, or does the introduction of multiple pours change the fundamental way in which molten copper interacts with the mold in which it is cast?

Examination of the ingots cast in two consecutive hot pours corroborates the observation that sand molds allow gases to escape more readily, while clay molds more effectively trap them, since once again the sand-cast ingot contains the fewest pockets of trapped gases. However, this pattern did not hold true for ingots cast in two pours where

the second pour of copper was made over an already cooled ingot. In this instance, the ingot cast in limestone had fewer gas pores than the ingot cast in sand. An ingot cast in a clay mold employing the cold two-pour method was not attempted because it was apparent that ingots produced in this manner looked nothing like those from the Uluburun shipwreck. Nevertheless, a discussion pertaining to the presence of pore distribution between the two ingots cast in this manner should be undertaken.

The distribution of pores between the cold double-pour ingots was drastically different and wholly unexpected. The sand-cast ingot contained pores only in the second pour of copper, while pores in the limestone-cast ingot exist only in and around the boundary between the two pour layers. These differing patterns of trapped gases are likely a direct result of the mold materials and their unique properties. What makes casting in multiple pours unique is that little or no gas is allowed to escape through the bottom of second pour of copper, as it is poured onto an already cooled layer of copper. This impenetrable floor of solid copper limits the viable avenues through which effervescing gases can escape to only the upper surface and the sides of the mold. One would think that these conditions would readily favor a sand mold for allowing more gases to escape, since a mold of sand is far more porous than one of limestone. However, this does not appear the case: why? The answer lies, it would seem, in the materials and manner in which the molds were constructed and their ability to retain the heat of molten copper. Based on observations concerning lack of gas pores, it appears that the ingot cast in limestone cooled much slower than did the ingot cast in sand, however this should not be the case. A mold of sand, with its pockets of air between grains of sand, should

actually be a better insulator than limestone mold, which would conduct heat away from the ingot. However, since the mold basin in the limestone mold was nearly twice as deep as the ingot was tall and the upper portion of the sand mold was actually a thin rim of sand, limestone proved to be better at insulating the cooling copper. The thin lip of sand that acted as the mold for the upper portions of the ingot allowed for the heat of the cooling ingot to be removed through convection, thus causing the sand-cast double-pour cold ingot to cool much quicker than its limestone counterpart, trapping more effervescing gases and creating a much more porous ingot.

Studying the microstructure of ingots, in combination with observations made by the unaided eye pertaining to their physical makeup, allows one to rule out certain mold materials as viable options for Late Bronze Age ingot casting technology. This hypothesis seems most convincing with respect to the use of sand as a viable mold material. Macroscopic examination of the single-pour ingot cast in sand reveals a grain structure unlike anything noted in Hauptmann et al.'s (2002) study and a physical structure too uniform to adequately compare to the ingots from the Uluburun wreck. For example, Hauptmann et al. (2002: 7) noted that the copper grains in the samples they studied were equiaxed; that is to say the copper grains were, on average, roughly the same size and shape throughout the copper sample. Sections of the single-pour ingot cast in sand, however, reveal a grain structure that is quite unlike what is described by Hauptmann et al. For instance, Figure 8 revealed highly directional grain structure characterized by many vertical columnar shapes. This feature is indicative of metal that cooled rapidly (Scott 1991: 6). Furthermore, Figure 25 shows copper grains that are not

equiaxed, as there is great disparity between the grain sizes and shapes. Arguments could be made to the effect that the columnar grain structure and disparity between grain sizes could be avoided, should the mold have been preheated. This thinking, however, can be easily refuted since a green sand mold can not be preheated because moisture is required for it to hold its shape. A green sand mold, in theory should have been a good insulator, causing any ingots cast therein to cool slower than those cast in limestone and clay. As discussed above with respect to the cold double-pour ingots, this was not the case. It appears that since the sand mold was not dug as deep and those of clay and limestone, and, in fact, had a thin lip acting as the upper section of the mold basin, convection of the heat contained in the copper away from the ingot caused it to cool quicker than any cast in limestone or clay.

Additionally, microscopic examination also reveals that the ingot cast in sand has significantly less precipitates along the grain boundaries than those analyzed from the Uluburun shipwreck (Figure 25). Moreover, many of the precipitates that do appear in the sample are contained within the borders of each copper grain, rather than along them.

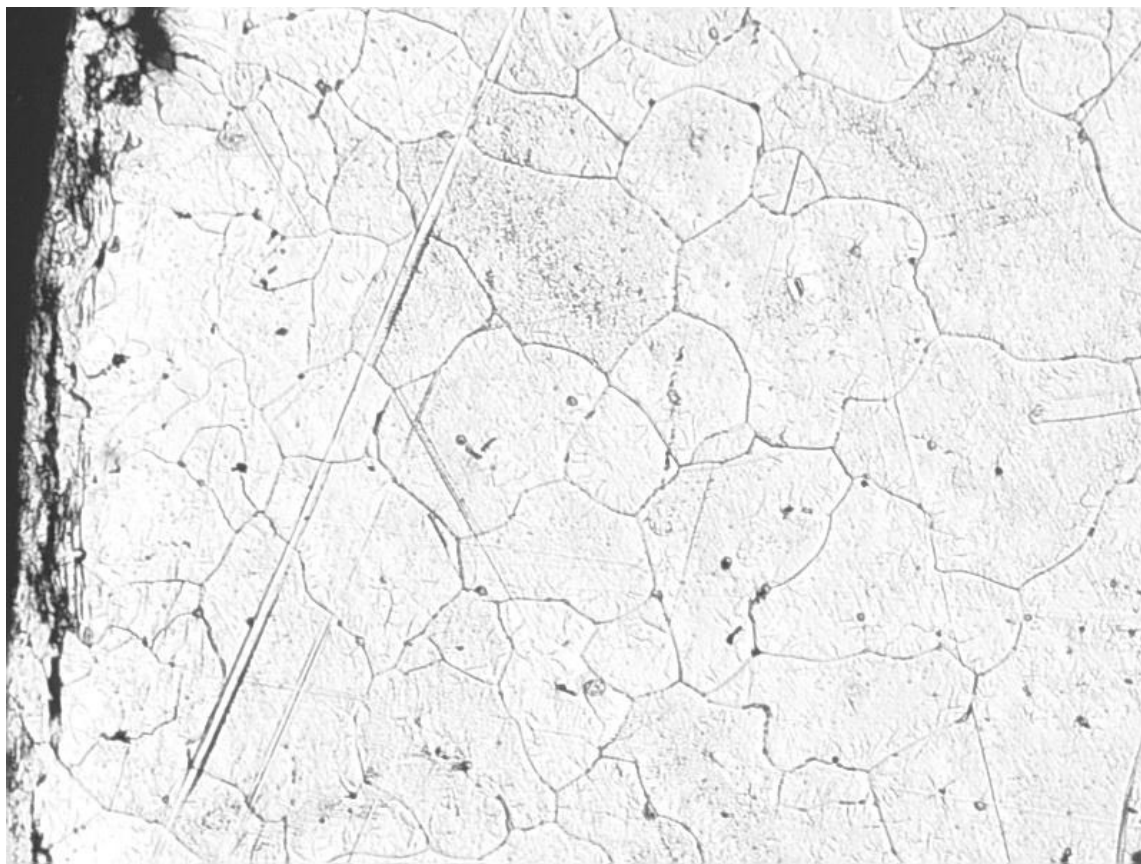


FIGURE 25. Clay mold, hot double-pour ingot at 200x magnification. Grains of dissimilar sizes with sporadic precipitates along the grain boundaries (photo by author, 2007).

With all the microscopic evidence against the use of green-sand molds as the likely mold material used for producing the Uluburun ingots, one should not overlook perhaps the most obvious and simplistic set of indicators that green-sand casting was probably not used in the Late Bronze Age, that is, the physical appearance of a sand-cast copper ingot itself. The ingot created in a green-sand mold with a single pour of copper is too uniform in shape and has surfaces that are far smoother than any observed in the assemblage of ingots from Uluburun. The rough surface of the green-sand cast ingot lacks the rough, bubbly texture of the Uluburun ingots. In fact, the rough surface is relatively flat and smooth and comprises of a thin film of metallic copper that is not like anything observed on any of the ingot from Uluburun. Possibly the most stark contrast between the ingot produced in these experiments and those from the Uluburun wreck are in the appearances of the mold surfaces. Uluburun ingots have many gas pores on their mold surfaces, while the ingot created in green sand has none. The lack of porosity on the green sand ingot is likely a function of the mold material, as sand would allow for the passage of escaping gases through the air pockets between individual sand grains that comprise the mold, rather than trapping them between the mold and the cooling ingot to form gas pockets in the mold and rough surfaces on the ingot.

Through microscopic analysis, it also appears there is good cause to rule out limestone as a possible mold material used in casting the Uluburun ingots, even though an ingot cast in this material bears many physical similarities to the ingots in the archaeological assemblage. The primary argument against limestone as a viable solution to the mold question stems from the lack of precipitates in the copper matrix. While the

copper grains in the ingot cast from a single pour of copper in a limestone mold are equiaxed in their structure and arrangement, there are too few precipitates along the grain boundaries when compared with those from the Uluburun wreck. As with the sand-cast ingot of a single pour, precipitates seem to occur with greater frequency inside the copper grains, rather than along their boundaries. In addition to the lack of precipitates, the single-pour ingot cast in limestone does not exhibit sufficient porosity at the microscopic level to be considered similar to the samples examined from Late Bronze Age copper ingots.

Admittedly, these results are somewhat puzzling given the observable physical characteristics of the ingots cast in limestone and their similarities to those ingots from Uluburun. The mold surface of the single-pour, limestone-cast ingot contains many gas pores that bear great resemblance to those found on the mold surfaces of nearly every ingot from Uluburun. In fact, of all the ingots cast in these experiments, no mold surface bears more similarities to the Uluburun ingots than that of the single-pour ingot cast in limestone.

Magnification does, however, reveal similarities between the ingots analyzed from the Uluburun wreck and the two single-pour ingots cast in clay molds. Comparisons between Figures 26 and 27 illustrate the granular similarities between these ingots. The porosity and accumulation of precipitates at grain boundaries within both ingots should be noted. Although grain sizes appear to differ slightly within the two ingots, the similarities are too great to be discounted. Preheating the mold prior to casting an ingot is a possible way to increase grain size of the copper. It is conceivable,

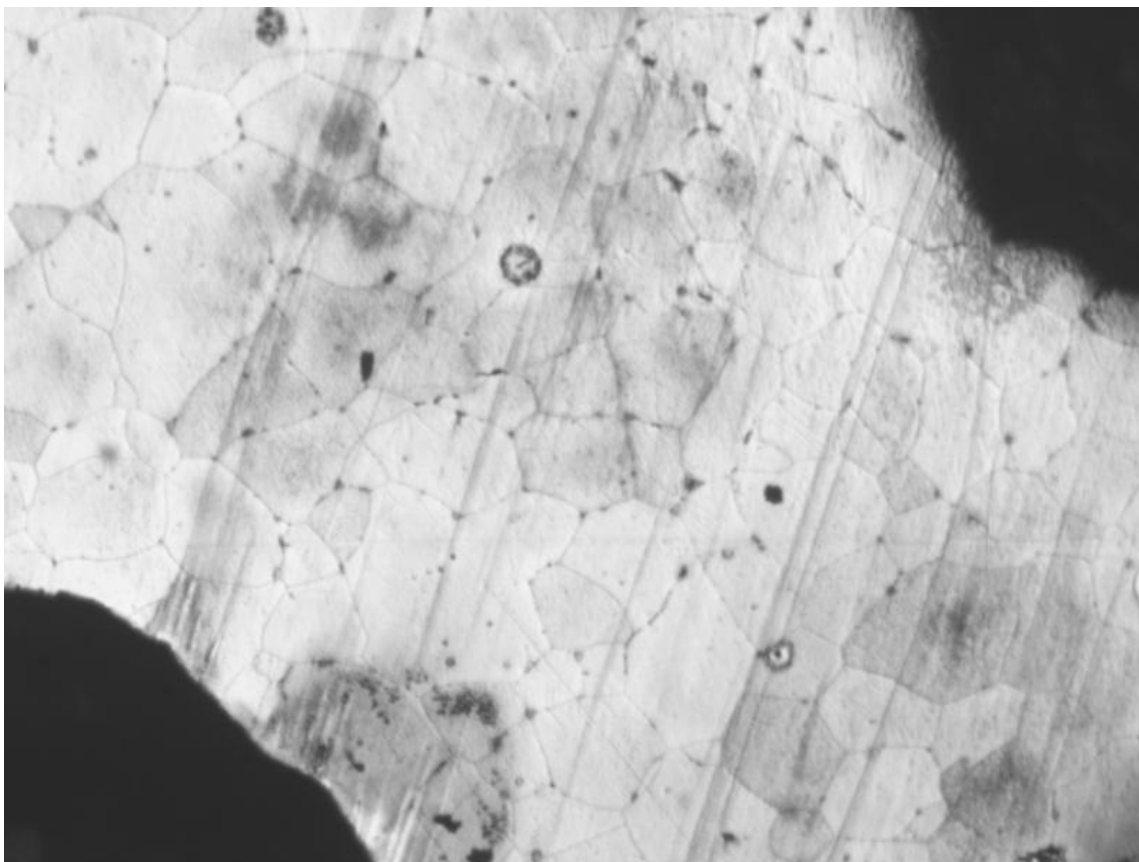


FIGURE 26. Grain structure of the single-pour ingot cast in a clay mold (seen at 200x magnification), with minimal precipitates along the grain boundaries (photo by author, 2007).

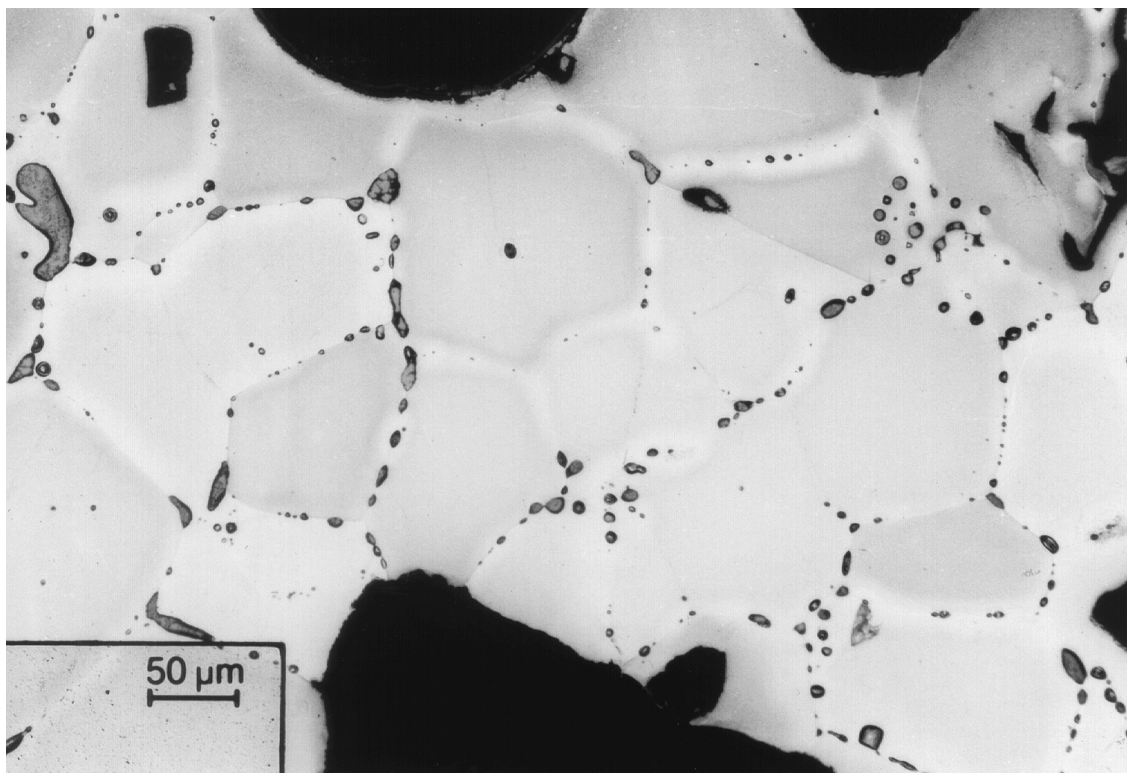


FIGURE 27. Grain structure of an Uluburun ingot. Note the similarities between the grain structure in this micrograph and those in Figure 26 (Hauptmann et al. 2002: 10).

should the clay mold have been preheated during these experiments, that the grain size would have been larger and closer in proportion to those of the ingots examined from the Uluburun wreck. Both the limestone and clay molds could have been preheated before use, although heating a green sand mold would have proved problematic because, unlike the other two molds, sand molds are not solid structures and rely on moisture to hold their shape.

On the basis of single-pour ingots produced in the trails and from a microscopic standpoint, it seems that clay is the most viable mold material for replicating the ingots from the Uluburun shipwreck. The single-pour ingot cast in limestone, on the other hand, seems to most closely resemble the archaeological ingots at a general level. However, while these conclusions may point toward some resolution in the casting query, there still remains the issue of the enigmatic striations around the perimeter of nearly every Uluburun ingot (Figure 28). These striations seem to allude to the idea that perhaps at least the large oxhide copper ingots in the Late Bronze Age were cast in multiple pours and that they represent the boundary between the first and second pours of copper. Castings ingots from two pours of copper were carried out in order to replicate these lines, but the results of those castings were inconclusive and the phenomenon behind these striations is still not fully understood.



FIGURE 28. A corner projection of an Uluburun oxhide ingot (KW 1069) suggesting that some of these ingots were cast in two pours in fairly rapid succession, resulting in a stacked arrangement of two separate ingots. (photo by author, 2004).

After preparing small sections of each experimental ingot for microscopic evaluation, it became readily apparent that the likelihood that the Uluburun ingots were produced by two separate pours of copper—the first pour being made onto an already cooled ingot—was extremely low. There are three reasons behind this assertion. The first and most obvious cause to dismiss the cold two-pour casting scenario is rooted in the ingots' physical appearance. When viewing each of the two experimental ingots formed by this method, it is overly apparent that they were produced by pouring molten metal over a cold surface, as evidenced by the sharp line delineating the two pours on the side of both ingots, and the fact that the second pour of copper overlapped the first in the limestone mold. While many ingots from the Uluburun shipwreck do show some type of line on their sides, none demonstrate the overlapping characteristics just described. In fact, only one (KW 2761) out of the 475 oxhide and plano-convex discoid ingots provides compelling evidence to make a case for two pours separated by sufficient time for the cooling of the first pour. The rough surface of a corner projection on ingot KW 2761 (Figure 29) is broken and reveals what appears to be another rough surface underneath, lending support to the idea of two pours separated by a long cooling period. However compelling this evidence may seem in support of two-pour ingots, where the second pour was made after the first formed its own rough surface, the division of the proposed two pours of KW 2761 still are not as pronounced as the two-pour ingots created in these experiments.



FIGURE 29. A damaged corner projection of one Uluburun oxhide ingot (KW 2761) supports the view that some of the ingots were cast in multiple pours (photo by author, 2004).

Following the overwhelming physical evidence that speaks against this method of casting, is the lack of a bond fusing the two pours of copper to one another. When sectioning the cold two-pour ingots for microscopic examination, the two halves representing the two pours separated into individual pieces in both instances; this did not occur during the preparation of any of the Uluburun ingot samples. While initially the two halves of the cold two-pour ingots appeared well-fused and integrated, indicating that perhaps the second pour of copper was hot enough to remelt the rough surface of the first pour and bond the two halves together, this was clearly not the case. It seems that instead of melding together with the first pour to create a single ingot, the second pour of copper seeped into the irregular surfaces of the first pour, creating a weak, interlocking, mechanical bond to the first pour that could be separated easily when the ingot was sample for sectioning purposes.

The final reason why it is unlikely that the Uluburun ingots were cast in multiple pours of copper, where the second pour was made on top of an already cooled ingot, was revealed through microscopic analysis. As discussed, the cold two-pour ingots were, in actuality, two separate ingots loosely held together and not the single solid unit as they initially appeared. This physical separation between halves is made even more apparent at the microscopic level, where it is clear that the uppermost copper grains of the first pour of copper are fused to second pour.

These observations lead to even more confusing aspects concerning the production of Late Bronze Age copper ingots. Clearly, on the basis of these experiments it can be said that the Uluburun ingots were not the product of a casting process that

involved an extended cooling period between successive pours of copper. This does not, however, rule out the possibility that the ingots were cast by multiple pours of copper made at sufficiently close intervals so that the initial pour did not have time to cool significantly. However, attempts to replicate that method of casting did not yield anything resembling the enigmatic striations that encompass many of the ingots from the Uluburun assemblage. At this time, it would seem that these striations are more a function of molten copper cooling and stacking on top of itself as it approached the edges of the mold, a phenomenon discussed at length by Whittick (1961) in his study of Romano-British lead ingots.

Since two separate experimental ingots were cast by two consecutive pours in the limestone mold, some observations and comments, however limited in scope they may be, can be made regarding the development of mold siblings. Mold sibling is a term used to group ingots together that appear to have been cast in succession in the same mold. Pulak, after careful examination, has revealed the presence of many mold siblings among the plano-convex discoid ingots from the Uluburun shipwreck (Pulak, 2000: 141).

Two successive pours into a limestone mold yielded interesting results with respect to the negative impressions of the mold left on the lower surfaces of the ingots. It has been noticed with the mold siblings from the Uluburun shipwreck that some of the diagnostic features, such as swells, on the mold surfaces tend to evolve along a continuum of prominence. That is to say a specific well on an ingot is larger and more pronounced than a corresponding swell on another ingot. This has been interpreted as the

ingot with the larger swell being cast later than the one with the corresponding smaller swell. Continuing with this example, the additional prominence of the swell was due to subsequent spalling and breakdown of the mold with each cast. However, these experiments suggest that the exact opposite may be the case.

Based solely on the appearances of the two ingots successively cast in the same limestone mold, it seems that the more prominent features are actually found on the first castings from a particular mold. Examination of the mold surfaces of these two ingots clearly shows that the shared mold surface features are sharper and more clearly defined on the first ingot cast in the mold, directly contradicting earlier predictions. After observing the breakdown of a limestone mold from the casting of a copper ingot, the explanation for this observation becomes apparent. As previously discussed and noted, limestone breaks down at 900° C, with the decalcining reaction increasing as temperatures exceed 1000° C. The results of that breakdown are not simply the spalling of the stone surfaces that are exposed to molten copper, but rather the surfaces turning to lime dust as well. Sharply defined features resulting from the carving of the mold would then be dulled and diminished as the mold surfaces turn to powder, thus generating less prominent features on the next ingots cast in that mold. Consequently, the detail of the diagnostic features used to establish the presence of mold siblings among a group of ingots, will, in reality, diminish with every ingot cast in a mold of limestone.

While it cannot be established with certainty, the work detailed in this thesis allows us to better understand what mold materials and the method of casting were used in the producing the Uluburun shipwreck ingots, as well as those produced elsewhere in

the Late Bronze Age. Based on this research it is not unreasonable to make certain sound conclusions concerning the origins of the Uluburun ingots. For example, because of the smooth and uniform physical shape, the columnar grain structure, and lack or precipitates along grain boundaries in sand cast ingots, it can be reasonably concluded that sand molds were not used when casting the Uluburun ingots. Furthermore, given the sharp division in the ingots between the first and second pours of copper, it can be argued that the Uluburun ingots were not cast in two successive pours of copper, regardless of mold material used, where the second pour was made onto an already cooled ingot surface. Outside of these two general observations, arguments for the methods of casting copper ingots in the Late Bronze Age become more muddled and less reliable.

Nevertheless, what has been established in the course of this research is a baseline to which all other ingots can be compared. The study outlined here seems to point toward clay as the likely mold material of choice for copper ingot casting in the Late Bronze Age, although this has not been proven. The study should also provide future researchers with a good starting point for delving further into this area of inquiry, in that it details the effects mold materials have on cooling copper, both on the macroscopic and microscopic levels. Indeed, while much has been learned about how copper interacts with clay, limestone, and sand molds, more research into the study of mold materials is needed. Preheating molds prior to casting also needs to be explored, when examining the effects of different methods used in the production of copper ingots. Furthermore, if sufficiently large furnaces can be made or utilized, and sufficient

quantities of matte copper can be procured, casting full-sized oxhide ingots would also be beneficial to gaining a more complete understanding of how copper smelters produced copper ingots in the Late Bronze Age.

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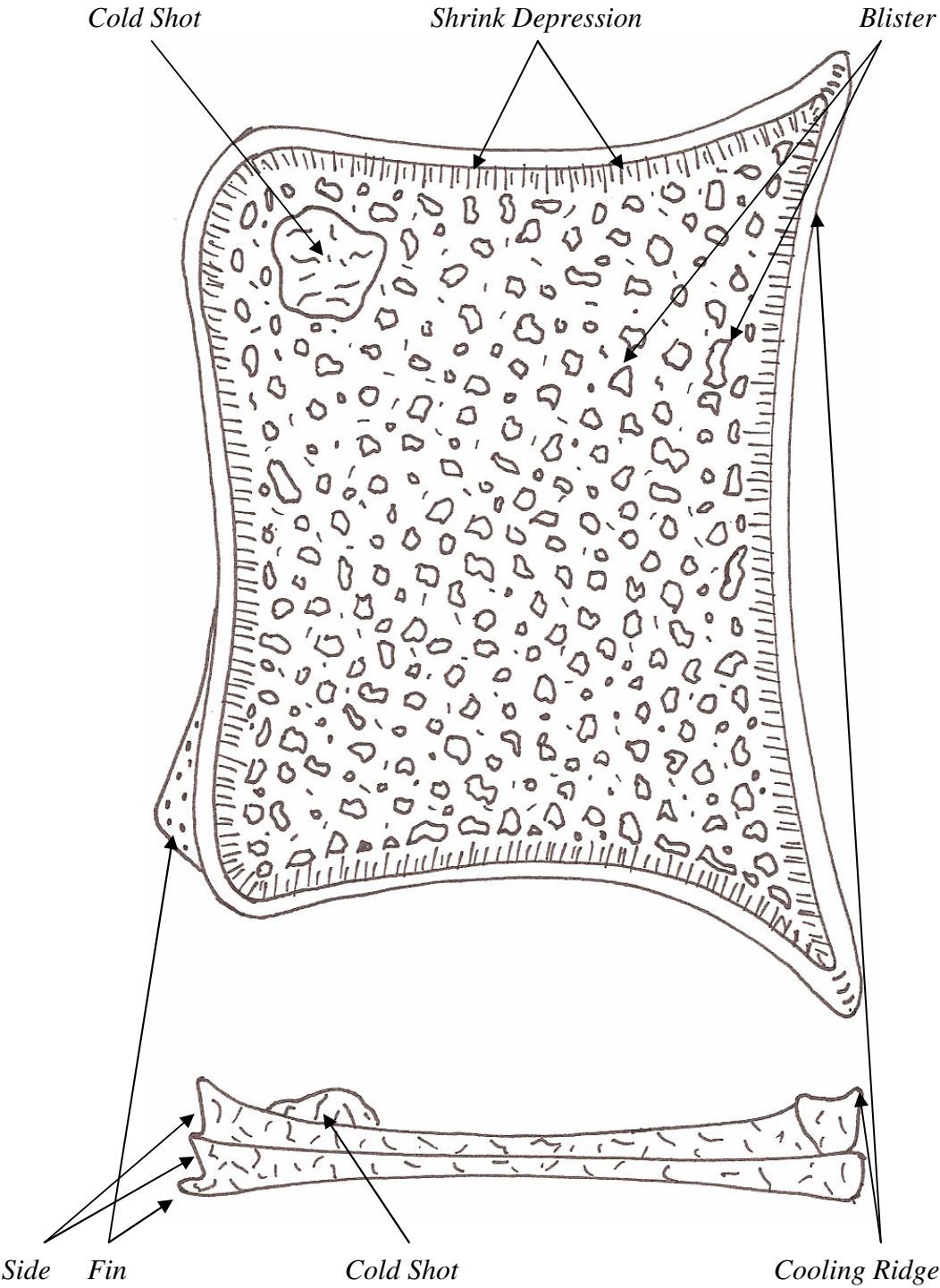
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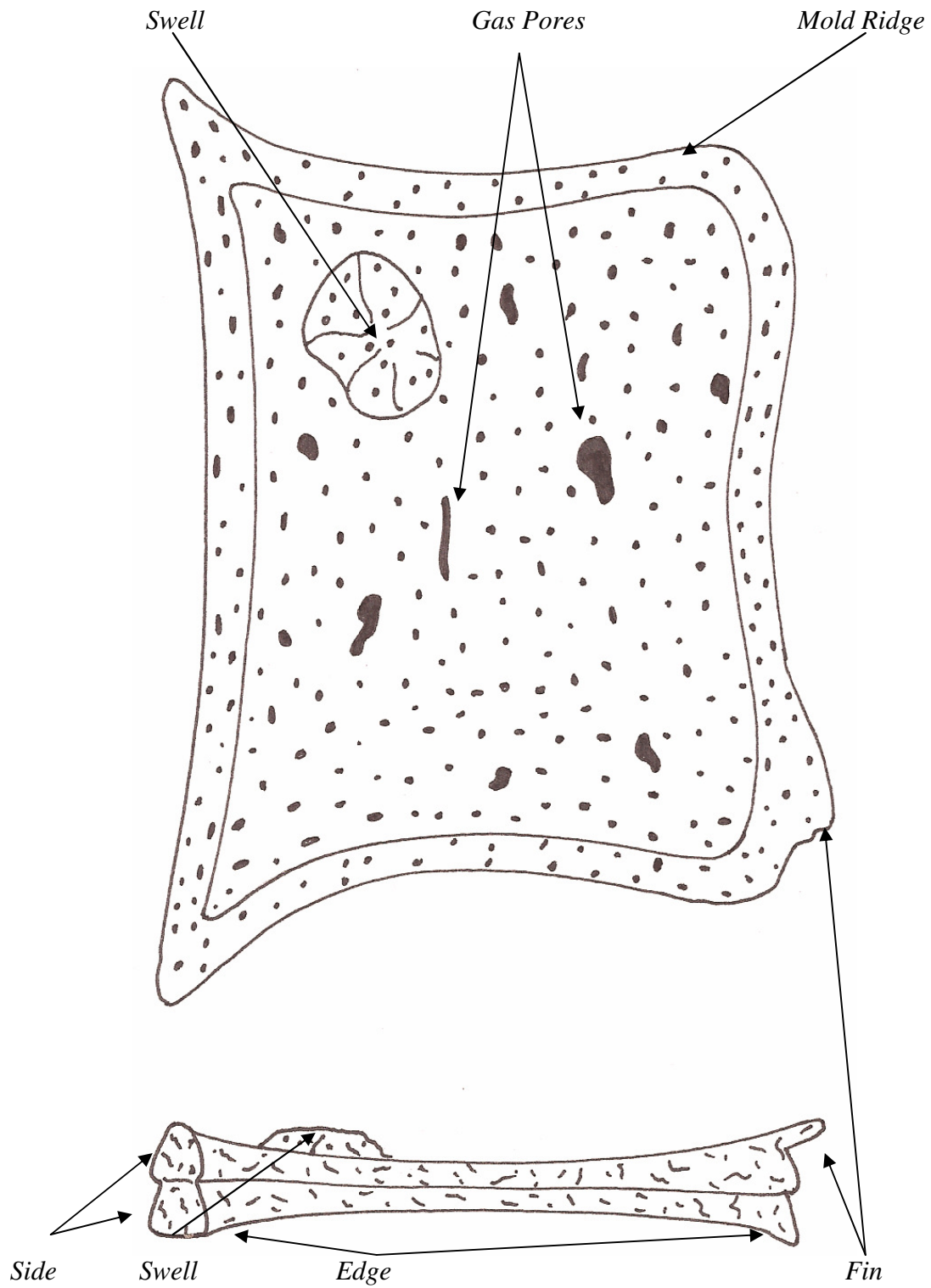
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APPENDIX A

Idealized Rough Surface of a Hypothetical Copper Oxide Ingot



Idealized Mold Surface of a Hypothetical Copper Oxide Ingot

APPENDIX B

Glossary of Casting Defect Terms

Blister – A shallow blow (round or elongated holes generated by trapped gas) covered over with a thin film of metal

Blister Ingot – Ingot cast of impure metal, marked by blisters and other casting defects.

Bun Ingot – Term used to define a small, round ingot that resembles a bun; synonymous with plano-convex discoid ingot.

Cold Shot – A freezing of the metal during pouring that results in a lump or ball resembling a metal shot on the objects rough surface.

Cooling Ridge – Ridge along the inside of the edge on the rough surface of an ingot that is caused by metal on the outer edge solidifying and creating a wall for which molten metal piles up against.

Equiaxed Grains – Grains that are approximately equal in all directions when comprising a metallic matrix.

Fin – A fin of metal on the casting caused by a crack in the mold or cope, etc.

Gas Pore – Widely dispersed round holes caused by gasses being absorbed in the metal during melting and then unable to be released during the solidifying of the casting.

Grain – The composite particles of a piece of metal; the arrangement of which determines the metals characteristics, such as hardness and texture.

Mold Surface – Surface on an ingot in contact with the mold during casting.

Mold Ridge – Ridge along the inside edge of an ingot on the mold surface. Unlike a cooling ridge, it is thought that this ridge is a result of the mold shape, not the early solidification of the edges of the ingot.

Oxhide Ingot – A flat, rectangular ingot cast of unrefined metal, mostly of copper, used during the Late Bronze Age. Protrusions at each corner and a slight narrowing at its middle gives the appearance of an outstretched ox hide, hence its name.

Oxidic – A compound of oxygen and another element.

Plano-Convex Discoid Ingot – Term used to define a circular ingot that is flat on one surface and rounded on the opposite surface, resembling a bun.

Precipitate – Microscopic non-metallic solids formed during the casting process found imbedded in the copper matrix.

Rough Surface – Surface of an ingot exposed to the atmosphere during casting.

Shrink Depression – A feature caused by a lack of feed metal causing a depression on the surface of the casting; a concave surface.

Swell – A casting deformation due to pressure of the molten metal moving or displacing part of the mold.

Tuyere - Clay tube, nozzle or pipe through which air is forced into a blast furnace to facilitate combustion.

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